

Aircraft engine exhaust emissions and other airport-related contributions to ambient air pollution

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6 **AIRCRAFT ENGINE EXHAUST EMISSIONS**
7 **AND OTHER AIRPORT-RELATED**
8 **CONTRIBUTIONS TO AMBIENT AIR**
9 **POLLUTION: A REVIEW**
10

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24 **Highlights**

- 25 ➤ Aviation is globally growing (+5% y^{-1}) mainly driven by developing countries
- 26 ➤ Airport operations cause an increase in ground-level pollution
- 27 ➤ Chemical and physical properties of the emitted gases and particles are reviewed
- 28 ➤ An overview of other additional sources within airports is provided
- 29 ➤ Future research needs on aircraft emissions are highlighted

30

31 **ABSTRACT**

32 Civil aviation is fast-growing (about +5% every year), mainly driven by the developing economies
33 and globalization. Its impact on the environment is heavily debated, particularly in relation to
34 climate forcing attributed to emissions at cruising altitudes and the noise and the deterioration of air
35 quality at ground-level due to airport operations. This latter environmental issue is of particular
36 interest to the scientific community and policymakers, especially in relation to the breach of limit
37 and target values for many air pollutants, mainly nitrogen oxides and particulate matter, near the
38 busiest airports and the resulting consequences for public health. Despite the increased attention
39 given to aircraft emissions at ground-level and air pollution in the vicinity of airports, many
40 research gaps remain. Sources relevant to air quality include not only engine exhaust and non-
41 exhaust emissions from aircraft, but also emissions from the units providing power to the aircraft on
42 the ground, the traffic due to the airport ground service, maintenance work, heating facilities,
43 fugitive vapours from refuelling operations, kitchens and restaurants for passengers and operators,
44 intermodal transportation systems, and road traffic for transporting people and goods in and out to
45 the airport. Many of these sources have received inadequate attention, despite their high potential
46 for impact on air quality. This review aims to summarise the state-of-the-art research on aircraft
47 and airport emissions and attempts to synthesise the results of studies that have addressed this issue.
48 It also aims to describe the key characteristics of pollution, the impacts upon global and local air
49 quality and to address the future potential of research by highlighting research needs.

50

51 **Keywords:** Aviation; atmospheric pollution; emissions; LTO cycles; particulate matter; oxides
52 of nitrogen

53

54 **List of abbreviations**

55	AAFEX	Alternative Aviation Fuel Experiment
56	AEs	Airport emissions
57	APEX	Aircraft Particle Emissions eXperiment
58	APU	Auxiliary power unit
59	BC	Black carbon
60	C*	Effective saturation concentration
61	CI_s	Chemi-ions
62	CIMS	Chemical ionisation mass spectrometry
63	EC	Elemental carbon
64	EI	Emission index
65	EXCAVATE	EXperiment to Characterise Aircraft Volatile Aerosol and Trace-species Emissions
66	F₀₀	Engine thrust expressed as a percentage of maximum rated power
67	FGEP	Fixed ground electrical power
68	FSC	Fuel sulfur content
69	FT	Fischer-Tropsch fuel
70	GMD	Geometric number mean diameter
71	GPUs	Ground power units
72	GRPs	Ground running procedures
73	GSEs	Ground service equipments
74	ICAO	International Civil Aviation Organization
75	LTO	Landing and take-off cycle
76	OC	Organic carbon
77	NMHC	Non-methane hydrocarbon
78	NO_x	Nitrogen oxides (NO+NO ₂)
79	NO_y	Reactive odd nitrogen (NO _x and their oxidation products)

80	OA	Organic aerosol
81	PAHs	Polycyclic aromatic hydrocarbons
82	PM	Particulate matter
83	PM₁	Particulate matter (aerodynamic diameter less than 1 µm)
84	PM_{2.5}	Particulate matter (aerodynamic diameter less than 2.5 µm)
85	PM₁₀	Particulate matter (aerodynamic diameter less than 10 µm)
86	RF	Radiative forcing
87	RPK	Revenue passenger kilometres
88	RTK	Revenue tonne kilometres
89	SARS	Severe acute respiratory syndrome
90	SIA	Secondary inorganic aerosol
91	SN	Smoke number
92	SOA	Secondary organic aerosol
93	SVOCs	Semi-volatile organic compounds
94	TC	Total carbon
95	TF	Turbofan engine
96	TIM	Time-in-mode
97	TJ	Turbojet engine
98	TP	Turboprop engine
99	TS	Turboshaft engine
100	UFP	Ultrafine particles (diameter <100 nm)
101	UHC	Unburned hydrocarbons
102	VOCs	Volatile organic compounds
103	ε	Abundance ratio ((SO ₃ +H ₂ SO ₄) /total sulfur)
104	ξ	Partitioning coefficient
105		

106 **1. INTRODUCTION**

107 Among pollution issues, poor air quality attracts a high level of interest within the scientific
108 community and engages public opinion because of the known relationship between exposure to
109 many air pollutants and increased adverse short- and long-term effects on human health (e.g.,
110 Schwartz, 1997; Ayres, 1998; Brunekreef and Holgate, 2002; Kampa and Castanas, 2008; Maynard,
111 2009; Yang and Omaye, 2009; Rückerl et al., 2011). In addition, air pollution can seriously impair
112 visibility (Hyslop, 2009), may damage materials in buildings and cultural heritage (Watt et al.,
113 2009; Screpanti and De Marco, 2009) and has direct and indirect effects upon climate (Ramanathan
114 and Feng, 2009). While air pollution remains a major concern for developing countries (Fenger,
115 2009; Liaquat et al., 2010) as a result of the rapid growth of population, energy demand and
116 economic growth, developed countries have experienced a significant decline in the concentrations
117 of many air pollutants over the past decade.

118
119 Airport emissions (AEs) have received increasing attention in recent years because of the rapid
120 growth of air transport volumes and the expected expansion to meet capacity needs for future years
121 (Amato et al., 2010; Kurniawan and Khardi, 2011; Kinsey et al., 2011). Most studies highlight
122 knowledge gaps (e.g., Webb et al., 2008; Wood et al., 2008a; Lee et al., 2010) which are a matter of
123 concern as the literature indicates that aircraft emissions can significantly affect air quality near
124 airports (Unal et al., 2005; Carslaw et al., 2006; Herndon et al., 2008; Carslaw et al., 2008;
125 Mazaheri et al., 2009; Dodson et al., 2009) and in their surroundings (Farias and ApSimon, 2006;
126 Peace et al., 2006; Hu et al., 2009; Amato et al., 2010; Jung et al., 2011; Hsu et al., 2012). Emission
127 standards for new types of aircraft engines have been implemented since the late 1970s by the
128 International Civil Aviation Organization (ICAO) through the Committee on Aircraft Engine
129 Emissions (CAEE) and the subsequent Committee on Aviation Environmental Protection (CAEP).
130 One of the key actions of the ICAO committees was the provision on engine emissions in Volume
131 II of Annex 16 to the Convention on International Civil Aviation, the so-called “Chicago

Convention”, which recommended protocols for the measurement of carbon monoxide (CO), nitrogen oxides ($\text{NO} + \text{NO}_2 = \text{NO}_x$), unburned hydrocarbons (UHC) and smoke number (SN) for new engines (ICAO, 2008). Standards were listed on a certification databank (EASA, 2013), which represents a benchmark for engine emissions performance and is used in many regulatory evaluations (ICAO, 2011). This regulation has produced significant improvements in engine and fuel efficiency and technical progress to reduce emissions. However, although these efforts have led to a substantial reduction in direct aircraft emissions over the past two decades, these gains may be offset by the forecast growth of the aviation industry and the resulting increase in airport traffic (ICAO, 2011). Furthermore, the ICAO regulation address only four main generic pollutants and a more detailed chemical and physical characterization of exhausts is required to quantitatively and qualitatively assess aircraft emissions. An increasing number of studies provide a detailed chemical speciation for many exhaust compounds, including gases and airborne particulate matter (e.g., Anderson et al., 2006; Herndon et al., 2008; Agrawal et al., 2008; Mazaheri et al., 2009; Onash et al., 2009; Herndon et al., 2009; Kinsey et al., 2011; Mazaheri et al., 2011; Santoni et al., 2011). However, the literature remains very sparse and many questions remain unresolved because of the large differences in measurement strategies, technologies and methods, compounds analysed and environments studied.

Aircraft exhausts are only one of several sources of emission at an airport (ICAO, 2011). Although exhaust plumes from aircraft engines were conventionally considered to account for most of the emissions, other sources are present within modern airports and contribute to air pollution at the local scale. Among these, tyre, brake and asphalt wear and the re-suspension of particles due to the turbulence created by the aircraft movements can account for large fractions of total particulate matter mass (e.g., British Airports Authority, 2006), but their chemical and physical characteristics have been investigated in only a few studies (Bennett and Christie, 2011; Bennett et al., 2011). Moreover, the emissions of the units providing power to the aircraft on the ground have received

158 relatively little consideration despite their potentially high impact on the local air quality (Schäfer et
159 al., 2003; Ratliff et al., 2009; Mazaheri et al., 2011). These units include the auxiliary power units
160 (APUs), which are small on-board gas-turbine engines, and the ground power units (GPUs)
161 provided by airports. In addition, airport ground service equipment (GSEs) further impact the air
162 quality (e.g., Nambisan et al., 2000; Amin, 2001; Schäfer et al., 2003). GSEs include most of the
163 equipment that an airport offers as a service for flights and passengers and includes a large number
164 of vehicles, such as passenger buses, baggage and food carriers, container loader, refilling trucks,
165 cleaning, lavatory services and de/anti-icing vehicles, and tugs, which are used to move any
166 equipment or to push the aircraft between gates and taxiways. Only few studies are available on the
167 air traffic-related emissions produced by ground services such as GSEs, GPUs or APUs (e.g., Webb
168 et al., 2008; Ratliff et al., 2009; Mazaheri et al., 2011; Presto et al., 2011).

169

170 Additional sources may also be present at airports, including maintenance work, heating facilities,
171 fugitive vapours from refuelling operations, kitchens and restaurants for passengers and operators,
172 etc. Moreover, as many airports are located far from cities, their emission inventories should also
173 include sources not directly present within a terminal, but on which the airport has an influence.
174 These sources may include intermodal transportation systems or road traffic including private cars,
175 taxis, shuttle buses and trucks for transporting people and goods in and out of the airport.

176

177 As most large airports are located near heavily populated urban settlements, in combination they
178 have a potentially significant impact on the environment and health of people living in their
179 vicinity. For example, 150 airports in the USA are located in areas designated to be in non-
180 attainment for one or more criteria air pollutants (Ratliff et al., 2009). In undertaking air quality
181 assessments and the development of successful mitigation strategies, it is therefore fundamental to
182 consider all the aspects associated with the entire “airport system”. However, current information
183 on many aspects of this polluting source is inadequate, including a detailed speciation of

184 hydrocarbons, physicochemical characteristics of particles, volatile and semi-volatile emissions and
185 especially the secondary transformations from the aging of aircraft exhausts and other airport-
186 related emissions. Some of these gaps are well summarised in a US Transportation Research Board
187 report (Webb et al., 2008).

188

189 **1.1 Aims and Outline of the Review**

190 Since the scientific literature on AEs remains very sparse and many questions are still open, this
191 review aims to summarise the state-of-the-art of airport emissions research and attempts to
192 synthesise and analyse the published studies. An overview of current information on airport-related
193 emissions is presented and the key characteristics of the pollution and the impacts on the local and
194 global air quality are discussed. This review further summarises the various methodologies used for
195 measurements and attempts to critically interpret the data available in the literature. Finally, this
196 review will highlight priority areas for research.

197

198 The next section traces the main stages of the development of civil aviation, by focusing especially
199 on the changes and development strategies of modern airport systems. Recent traffic data and
200 statistics are presented and the trends are also discussed in order to understand the potential future
201 growth of air transport, which is fundamental to forecasting the impacts of aviation in future years.
202 The third section gives an overview of the operation of aircraft engines, briefly discusses the most
203 widely used technologies, describes some fuel characteristics, such as the sulfur content, and
204 analyses the current use and future jet fuel consumption scenarios. The fourth section reviews the
205 current information on aircraft engine exhaust: the landing and take-off cycles are described since
206 they are commonly used to assess aircraft emissions during the operational conditions within an
207 airport and within the atmospheric surface boundary layer; the main gaseous and particulate-phase
208 compounds emitted by aircraft are listed and their key chemical and physical characteristics are
209 described in separate subsections. A summary of data on the emission indices for many pollutants is

210 also provided. The fifth section describes the non-exhaust emissions related to aircraft operations,
211 such as the tyre and brake wear and the re-suspension of runway material, which have been little
212 investigated even though they may have serious impacts on local air quality. The sixth section
213 reviews data on the non-aircraft emissions potentially present within an airport, including the
214 ground service equipment emissions, the auxiliary/ground power units and others. The seventh
215 section presents the results of studies conducted indoors and outdoors at airports to directly assess
216 the impacts of AEs upon human health. Finally, this paper reviews the results of the recent literature
217 on aircraft emissions and other airport-related contributions to highlight the potential role of AEs
218 upon local air quality.

219

220 **2. PRESENT SCENARIOS AND FUTURE PERSPECTIVES OF CIVIL AVIATION**

221 **AND AIRPORTS**

222 The Airport Council International (ACI, 2013) has reported recent statistics on the air traffic
223 volumes for 2012: more than 79 million aircraft movements carried annually 5.7 billion passengers
224 between 1,598 airports located in 159 countries, and reported that the total cargo volume handled by
225 airports was 93 million tonnes. However, these numbers are expected to further increase in the
226 forthcoming decades: in the past half century, the aviation industry has experienced a strong and
227 rapid expansion as the world economy has grown and the technology of air transport has developed
228 (Baughcum et al., 1999). Generally, air traffic has been expressed as revenue passenger kilometres
229 (RPKs) by multiplying the number of revenue-paying passengers aboard the vehicle by the travelled
230 distance, or occasionally in revenue tonne kilometres (RTK). Figure 1 shows the absolute growth of
231 aviation recorded by ICAO in terms of RPK, RTK and aircraft kilometres from the 1930s to today
232 (ICAO, 2013; Airlines for America, 2013). Despite some global-scale events, such as the Gulf crisis
233 (1991), the terrorist attack of 11th September 2001, the outbreak of severe acute respiratory
234 syndrome (SARS) in 2002–2003 and the recent global economic crisis (2008–2009), an average
235 annual growth rate of 5% was observed and this trend is expected to continue over the next decades

236 mainly driven by the economic growth of emerging regions (ACI, 2007; 2008; Airbus, 2012;
237 Boeing, 2013). It is anticipated that there will be more than 9 billion passengers globally by 2025
238 and more than 214 million tonnes of total world freight traffic are forecast over almost 120 million
239 air traffic movements (ACI, 2007). The future growth of air transport will inevitably lead to the
240 growth of airline fleets and route networks and will therefore lead to an associated increase in
241 airport capacity in terms of both passengers and cargo. This poses questions as to the consequent
242 impact on air quality.

243

244 **3. AIRCRAFT: CHARACTERISTICS AND IN-USE TECHNOLOGIES**

245 Emissions from aircraft engines are recognised as a major source of pollutants at airports and have
246 been extensively investigated over the past 40 years. Initially, the main historical concern for
247 supersonic aircraft was over stratospheric ozone depletion (Johnston, 1971) and secondarily about
248 the formation of contrails at cruising heights (Murcray, 1970; Schumann, 2005) and indirect effect
249 on the Earth's radiative budgets (Kuhn, 1970). Apart the development of the Concorde and the
250 Tupolev Tu-144, a supersonic fleet flying in the stratosphere was never developed and today all
251 commercial airliners are subsonic equipped with turbofan or turboprop engines. Therefore, the main
252 present issue arising from civil aviation has today shifted to the increased levels of ozone in the
253 upper troposphere and lower stratosphere resulting from the atmospheric chemistry of emitted NO_x
254 (Lee et al., 2010 and reference therein). Furthermore, the development of increasingly restrictive
255 legislation on ambient air quality and the implementation of enhanced monitoring networks in many
256 developed countries has highlighted the effects of aircraft emissions at ground-level and the
257 deterioration of air quality near airports.

258

259 **3.1 Engines**

260 Engines for civil and general aviation are generally classified as gas turbine engines (turbofan and
261 turboprop) fuelled with aviation kerosene (also named jet fuel) and internal combustion piston

262 engines fuelled with aviation gasoline, often referred as avgas (ICAO, 2011). The majority of
263 modern airliners are equipped with turbofan engines. These engines are derived from predecessor
264 turbojet engines developed during World War II. A turbojet is composed of an inlet compressor, a
265 combustion section adding and igniting fuel, one or more turbines extracting energy from the
266 exhaust gas in expansion and driving the compressor. A final exhaust nozzle accelerates the exhaust
267 gas from the back of the engine to generate thrust. Turbofan engines use a turbojet as a core to
268 produce energy for thrust and for driving a large fan placed in front of the compressor. In modern
269 airliners, the fan provides most of the thrust. The “bypass ratio” refers to the ratio of mass flux
270 bypassing the combustor and turbine to the mass flux through the core: high-bypass ratios are
271 preferred for civil aviation for good fuel efficiency and low noise. Some small and regional airliners
272 are instead equipped with turboprop engines, which use a turbine engine core fitted with a reduction
273 gear to power propellers. A simplified diagram of a turbofan engine is provided in Figure 2. In
274 August 2013 the ICAO (EASA, 2013) listed a total of 487 in-use turbofan engines (including
275 packages): Table 1 provides a summary of the current engine families mounted in the most popular
276 airliners (75% of total in-use turbofan engines).

277

278 Reciprocating piston engines are predominately fitted in small-sized aircraft typically related to
279 private use, flying clubs, flight training, crop spraying and tourism. Internal piston engines run
280 under the same basic principles as spark ignition engines for cars, but generally require higher
281 performance. Four-stroke-cycle engines are commonly used, more rarely these can be two-stroke
282 and occasionally diesel. The principal difference between jet and piston engines is that combustion
283 is continuous in jet engines and intermittent in piston engines. Other flying vehicles may be present
284 within an airport, such as helicopters. These vehicles are usually less numerous than the airliners in
285 most terminals, but in some circumstances their contribution to the air quality cannot be
286 disregarded. Today, most modern helicopters are equipped with turboshaft engines, whose

287 functioning is similar to a turbojet but are optimised to generate shaft power instead of jet thrust.

288 This review abbreviates turbojet (TJ), turbofan (TF), turboprop (TP) and turboshaft (TS).

289

290 **3.2 Fuel Characteristics**

291 At the current time, almost all aviation fuel (jet fuel) is extracted from the middle distillates of crude
292 oil (kerosene fraction), which distils between the gasoline and the diesel fractions. The kerosene-
293 type fuels most used worldwide in civil aviation are of Jet A and Jet A-1 grades: Jet A is used in
294 most of the world, except North America where Jet A-1 is used. An exhaustive review of jet fuel
295 production processes is given elsewhere (Liu et al., 2013). The specifications of such fuels are
296 addressed by two organizations, the American Society for Testing and Materials (ASTM) and the
297 United Kingdom Ministry of Defence (MOD). Jet A is used for almost all commercial aviation
298 flying within or from the USA and is supplied against the ASTM D1655 specification. It has a
299 flash point minimum of 38°C and a freeze point maximum of -40°C. Jet A-1 is widely used outside
300 the USA and follows the UK DEF STAN 91-91 (Jet A-1) and ASTM D 1655 (Jet A-1)
301 specifications. It has same flash point as Jet A but a lower freeze point (maximum of -47°C) and a
302 mean C/H ratio of $C_{12}H_{23}$ (Lewis et al., 1999; Chevron Corporation, 2006; Lee et al., 2010). Other
303 fuels can be used as an alternative to Jet A-1. Jet B is a wide-cut type fuel covering both the naphtha
304 and kerosene fractions of crude oil and is used in very cold climates, e.g. in northern Canada where
305 its thermodynamic characteristics (mainly lower freeze point and higher volatility) are suitable for
306 handling and cold starting. ASTM publishes a specification for Jet B, but in Canada it is supplied
307 against the Canadian specification CAN/CGSB 3.23. Other specifications also exist such as
308 DCSEA (France) and GHOST (Russia). TS-1 is the main jet fuel grade available in Russian and
309 CIS states, along with T-1, T-2 and RT; it is a kerosene-type fuel with slightly higher volatility
310 (flash point is 28°C minimum) and lower freeze point ($\leq -50^\circ\text{C}$) compared to Jet A and A-1 fuels.
311 Various types of jet fuels are instead regulated by Chinese specifications: RP-1 and RP-2 are
312 kerosene-type fuels similar to Russian TS-1, while RP-4 to Jet B. Nowadays, virtually all jet fuel in

313 China is RP-3, which is quite comparable to Jet A-1 (Shell, 2013). Fuels for military purposes are
314 formulated for high-performances and are regulated separately by many governments; some of
315 these (JP grades for USA and NATO forces) were used in several studies (e.g., Anderson et al.,
316 2006; Chen et al., 2006; Cowen et al., 2009; Cheng et al., 2009; Cheng and Corporan, 2010;
317 Santoni et al., 2011). The kerosene-based JP-8 grade is currently the primary fuel for NATO
318 aircraft. Corporan et al. (2011) reported some JP-8 characteristics.

319

320 Jet fuels are a mixture of thousands of different hydrocarbons. The range of their molecular weights
321 is restricted by the distillation: in kerosene-type fuels (e.g., Jet A and Jet A-1) the carbon number
322 ranges between about 8 and 16, while in wide-cut jet fuels (Jet B), between about 5 and 15. Spicer
323 et al. (1994) reported that jet fuel is primarily composed of species with five or more carbons and
324 70% of the compounds by weight contain 11–14 carbon atoms. Most of the hydrocarbons in jet fuel
325 are members of the normal paraffins, iso-paraffin, cycloparaffin, aromatic and alkene classes: 20%
326 *n*-paraffins, 40% iso-paraffin, 20% naphthenes and 20% aromatics are typical (Lindstedt and
327 Maurice, 2000; Liu et al., 2013 and reference therein). Moreover, a series of different additives are
328 required or approved for use by ASTM and DEF STAN specifications to enhance or maintain some
329 fuel properties, improve performance or handling. Among those approved for Jet A and Jet A-1
330 fuels, some hindered phenols serve as antioxidants, the di-ethylene glycol monomethyl ether acts as
331 icing inhibitor, the N,N'-disalicylidene-1,2-propane diamine is added as chelating agent for many
332 metal ions. Other additives act as electrical conductivity/static dissipaters, corrosion inhibitor and
333 biocides: a summary is listed in Chevron Corporation (2006).

334

335 The aviation industry is nowadays investing significant effort towards the use of alternative fuels
336 (Blakey et al., 2011; Williams et al., 2012). Since aircraft emissions are recognised to be closely
337 linked to the fuel composition (Beyersdorf et al., 2013 and reference therein), recently the
338 introduction of synthetic fuels and bio-fuels instead of common oil-derivate jet fuels has been much

discussed in terms of beneficial effects upon exhaust emissions (e.g., Corporan et al., 2005; 2007; DeWitt et al., 2008; Timko et al., 2010a; Corporan et al., 2011; Lobo et al., 2011; Williams et al., 2012; Cain et al., 2013). Among others, the Fischer-Tropsch (FT) fuel seems to be a potential candidate for replacing, or mixing with, oil-derived conventional jet fuels. The FT reaction was developed in the first half of twentieth century and uses a mixture of carbon monoxide and hydrogen to produce a complex product stream of paraffins, olefins, and oxygenated compounds such as alcohols and aldehydes via product upgrading (e.g., cracking, fractionation, and isomerisation). The mechanism is explained in Liu et al. (2013). The FT process leads to a fuel with low aromatic content and no sulfur, which are reported to be beneficial in reduction of emissions of particulate matter and its precursors from aircraft engines (Corporan et al., 2007; Timko et al., 2010a; Lobo et al., 2011). Corporan et al. (2011) report gas chromatograms and hydrocarbon content of JP-8 and various alternative jet fuels. To study the effects of FT fuel usage on aircraft gaseous and particulate emissions the Alternative Aviation Fuel Experiment (AAFEX) was carried out in 2009: results are spread across various papers (e.g., Lee et al., 2011; Santoni et al., 2011; Anderson et al., 2011; Kinsey et al., 2012a,b; Beyersdorf et al., 2013).

Avgas for general aviation is distilled separately from the most common motor gasoline and is formulated for stability, safety, and predictable performance under a wide range of environments. Nowadays there are two main grades (100 and 100LL low lead) regulated by the ASTM D 910 and UK DEF STAN 91-90 specifications. Tetraethyl Pb is added to avgas for increasing fuel octane and avgas 100LL has a lead content up to 0.56 g Pb L^{-1} . The impact of general aviation is under discussion, since it was reported as one of the largest remaining source of lead emissions to the air in the USA (e.g., Carr et al., 2011). Avgas is principally composed of isoparaffinic and aromatic hydrocarbons and their carbon numbers vary from about 4 (butane) to 10, with the most prevalent carbon number being 8 (Chevron Corporation, 2006). It may include tetraethyl lead as antiknock additive, icing inhibitors, antioxidants and others.

365 **3.3 Sulfur Content in Fuels**

366 Over the past decades there has been a worldwide trend to decrease sulfur content in fuels and many
367 jurisdictions, including the USA and the European Union, have recently required very low sulfur
368 levels in road and marine fuels to reduce the SO_x and particulate matter emissions from the
369 transport sector. A similar reduction has not occurred for jet fuel although at the beginning of the
370 2000s the IPCC indicated that reducing the sulfur content of kerosene will reduce SO_x emissions
371 and sulphate particle formation (IPCC, 1999). The maximum sulfur content of aviation fuel has
372 remained at 3 g S kg fuel⁻¹, or 3000 ppm by mass (Lewis et al., 1999; Ebbinghaus and Wiesen,
373 2001; Anderson et al., 2005; Barrett et al., 2012). However, lower values of fuel sulfur content
374 (FSC) have commonly been reported: Fahey et al.(1999) stated that in the world market at the
375 beginnings of the 2000s the FSC was near 400 ppm; Hileman et al. (2010) reported that average
376 FSC in commercial Jet A, Jet A-1 and military JP-8 fuel grades varied between 550 to 750 ppm;
377 Agrawal et al. (2008) reported that FSC in the fuel was 300 ppm. Popovicheva et al. (2004) and
378 Demirdjian et al. (2007) reported that the aviation kerosene TS-1 has a FSC of 1100 ppm and less
379 than 10⁻⁴ wt.% of metals.

380

381 FSC in jet fuels is directly related to the SO₂ emissions in aircraft exhaust (e.g., Arnold et al.,
382 1998a; Schumann et al., 1998; Hunton et al., 2000). Some research projects, such as APEX-1, were
383 designed to study the effects of FSC on aircraft engine emissions (e.g., Wey et al., 2006; 2007;
384 Kinsey, 2009; Onash et al., 2009). Generally the studies reported that the emissions of both SO₂ and
385 sulphates are proportional to S levels in fuels, but no systematic difference between the low and
386 high sulfur fuels in terms of other emitted organic sulfur species (OCS and CS₂) were reported
387 (Anderson et al., 2006). The conversion of S(IV) to S(VI) is amply discussed later in this review.

388

389 Recently, the impact of ultra-low sulfur jet fuel (15 ppm) upon public health, climate, and
390 economics was examined by Barrett et al. (2012). They reported that the use of ultra-low sulfur

391 fuels on a global-scale will cost 1–4 billion US \$ per year, but may prevent 900–4000 air quality-
392 related premature mortalities per year. Moreover, Barrett and co-authors also stated that the
393 radiative forcing (RF) associated with reductions in atmospheric sulphate, nitrate, and ammonium
394 loading can be estimated as $+3.4 \text{ mW m}^{-2}$, i.e. equivalent to about 1/10th of the warming due to
395 CO₂ emissions from aviation.

396

397 **3.4 Current Use and Future Jet Fuel Consumption Scenarios**

398 The availability of reliable information on fuel consumption is essential to make robust estimates of
399 aviation emissions at both global and regional scales. Various estimates of aviation fuel
400 consumption are available in the literature and generally refer only to jet fuel, since piston-powered
401 flights were estimated to account for approximately 2% of propeller (piston plus turboprops) and ~
402 0.05% of total (propeller plus jet) fuel burn (Kim et al., 2007). Gauss et al. (2006) estimated a total
403 of 169 Tg fuel globally burned in 2000, of which 152 Tg is due to civil flights. The AERO2k global
404 aviation emissions inventories reported a total of 176 Tg of kerosene used in 2002 for both civil
405 (156 Tg) and military (19.5 Tg) aviation (Eyers et al., 2004); other studies of the 2000-2005 period
406 estimated that the global aviation industry consumed approximately 170-203 Tg of kerosene per
407 year with an evident decrease in 2001-2002 following the drop of aviation traffic due to the 11th
408 September 2001 and SARS events (Kim et al., 2007); Wilkerson et al. (2010), Whitt et al. (2011)
409 and Olsen et al. (2013) reported that the global commercial aircraft fleet burned 188 Tg of fuel in
410 2006; Chèze et al. (2011) reported a world consumption of 229 Mt of jet fuel in 2008. These
411 estimates accounted for approximately 3% of current annual fossil fuel energy usage (Barrett et al.,
412 2010, and reference therein). Data from OPEC (Mazraati, 2010) stated that the aviation sector in
413 2006 was the second major consumer of total oil demand in the transportation sector (11.2%) and
414 accounted for 5.8% of total oil consumed in the world. Given the past and future growth of the
415 aviation industry, this consumption may rise further: AERO2k emission inventories estimated a
416 forecast scenario for 2025 in which the fuel demand for aviation will be 327 Tg y⁻¹ (Eyers et al.,

2004); Chèze et al. (2011) reported that the world jet fuel demand is projected to grow by 38% between 2008 and 2025, rising to more than 316 Mt in 2025 at a mean growth rate of 1.9% per year. Owen et al. (2010) estimated the future global aviation emissions under four of the IPCC/SRES (Intergovernmental Panel on Climate Change/Special Report on Emissions Scenarios) marker scenarios and reported a fuel use of 336 Tg in 2020 and varying from 426 and 766 Tg for 2050. This study also reported an estimate of 325 Tg for 2050 if the ambitious technology targets of the Advisory Council for Aeronautical Research in Europe (ACARE, 2002) were to be achieved. Table 2 summarises the yearly global fuel consumption reported in recent studies. However, aviation traffic growth and jet fuel demand have been shown not to be strictly correlated, since the efficiencies of aircraft engines and air traffic management are improving and modern airliners are 75% quieter with consequent fuel consumption reduced by 70% with respect to the 1960s (Baughum et al., 1999; Nygren et al., 2009, and references therein). In particular, the current average fuel consumption of in-use fleets was estimated to be less than 5 L fuel every 100 RPK, while in most modern aircraft it drops to approximately 3.5 L / 100 RPK: Nygren et al. (2009) reported the historical world fleet of aircraft average fuel consumption and found an exponential trend in fuel consumption reduction from 1987 to the present day. Oil prices have driven investment in more efficient aircraft models. Fuel costs exceed those of labour costs for airlines. Fuel costs accounted for ~13% of total costs in 2002, but today they are closer to 34% (Boeing, 2013).

435

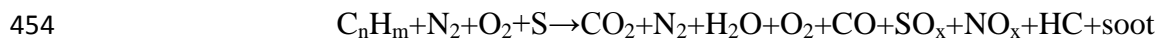
436 **4. AIRCRAFT EXHAUST EMISSIONS**

Emissions from aircraft engines are generally considered to be the dominant source at airports and the large majority of studies available in the literature focus on aircraft emissions. Common airliners burning kerosene-type fuels primarily produce carbon dioxide and water (Wahner et al., 1995; Lewis et al., 1999; Anderson et al., 2006; Lee et al., 2010), which are directly related to the burned fuel, with minor variations due to the carbon-hydrogen ratio of the fuel. In this context, it is

442 reported that the fuel flow of common airliner engines is approximately linearly proportional to
443 engine thrust setting (e.g., Anderson et al., 2005; Wey et al., 2006).

444

445 The oxidation of atmospheric nitrogen at the very high temperatures in engine combustors drives
446 the formation of nitrogen oxides, while the presence of trace amounts of sulfur, nitrogen and some
447 metals (e.g., Fe, Cu, Zn) in fuels (Lewis et al., 1999) and non-ideal combustion conditions within
448 engines may lead to the production of by-products, including sulfur oxides, additional nitrogen
449 oxides, unburned hydrocarbons and particulate soot. Furthermore, exhausts can also contain species
450 from the combustion and release of lubricant oils (Dakhel et al., 2007; Timko et al., 2010b; Yu et
451 al., 2010; Kinsey et al., 2011; Yu et al., 2012) and from mechanical component wear (Petzold et al.,
452 1998; Demirdjian et al., 2007). Therefore a more realistic, but simplified, combustion scheme in
453 aircraft engines can be summarised as (Lee et al., 2009):



455 IPCC reported that approximately 99.5-99.9% of the molar content of typical commercial engine
456 exhaust consists of N_2 , O_2 , CO_2 , and H_2O (Lewis et al., 1999). Figure 3 reports a more detailed
457 breakdown of combustion products for a core engine mass flow: the combustion products in aircraft
458 exhausts are mainly made up of CO_2 (~72%), H_2O (~27.6%), while residual products account for
459 less than 1%. Figure 2 summarises the main exhaust components of aircraft engines and their
460 potential effects on the environment and human health. It is estimated that roughly 90% of aircraft
461 emissions, except hydrocarbons and CO (~70%), are produced while cruising at altitude, while the
462 remainder is emitted during landing, take-off, and ground level operations (e.g., FAA, 2005).

463

464 Aircraft emissions have been studied extensively since the late-1960s and initially the interest was
465 mainly driven by their direct and indirect effects on climate and the generation of contrails. For this
466 reason, many early studies focused on emissions at high cruise altitudes (e.g., Reinking, 1968;
467 Kuhn, 1970; Arnold et al., 1992; Fahey et al., 1995a,b; Wahner et al., 1995; Brasseur et al., 1996;

468 Schumann, 1996;1997; Anderson et al., 1998a,b). The interest in aviation emissions at airports also
469 dates back many years (e.g., Daley and Naugle, 1979; Naugle and Fox, 1981), but only recently was
470 there an increasing awareness of the effects of aircraft emissions at ground level, or at least within
471 the planetary boundary layer. The recent interest in aircraft emissions at ground-level was initially
472 motivated by public concern, given that more and more often airports are held responsible for air
473 pollution and noise in nearby residential areas (e.g., Mahashabde et al., 2011). Since aircraft
474 emissions are related to engine thrust (e.g., Anderson et al., 2006; Lobo et al., 2007; Whitefield et
475 al., 2008; Timko et al., 2010b; Kinsey et al., 2010; Kinsey et al., 2011) and engines are designed for
476 high performance while cruising at high altitudes, some aircraft operations within airports require
477 that engines operate outside of their optimal regimes, ranging from maximum thrust during take-off
478 to low power settings during operations on the ground. This fact was clearly highlighted during the
479 APEX-1 campaign by Onash et al. (2009), who reported that a CFM56 engine is less efficient at the
480 low thrust levels usually used at airports. This may result in potentially higher emissions on the
481 ground than that during cruising for those pollutants mainly emitted at low power, such as CO and
482 hydrocarbons.

483

484 Early reports of nitrogen oxides, carbon monoxide, hydrocarbons and particulate matter from jet
485 aircraft turbine engines were made by Spicer et al. (1984). Subsequent studies (Spicer et al., 1992;
486 1994) added further information and provided detailed information on the organic component of
487 turbine engine emissions. Following from these pioneering studies, the scientific literature now
488 comprises a large number of studies and most have concluded that aircraft exhausts are responsible
489 for significant emissions of a series of gaseous, semi-volatile and non-volatile species. Non-volatile
490 emissions are produced in the combustor and are made up of refractory material such as soot (e.g.,
491 Agrawal et al., 2008; Kinsey, 2009; Dodson et al., 2009; Lee et al., 2010; Presto et al., 2011), which
492 is emitted into the atmosphere as particulate matter even at the high engine exit temperatures, but
493 also contains many organic compounds (e.g., Herndon et al., 2006; Anderson et al., 2006; Webb et

494 al., 2008; Wood et al., 2008a; Agrawal et al., 2008; Herndon et al., 2009; Lee et al., 2010; Mazaheri
495 et al., 2011; Presto et al., 2011; Kinsey et al., 2011; Mazaheri et al., 2013).

496

497 Volatile emissions include compounds that exists as vapour at engine exit temperature and pressure
498 (Presto et al., 2011) and are made up of gaseous and vapour-phase pollutants, such as CO₂, CO,
499 NO_x, SO₂, O₃ and many organic compounds, including alkanes, alkenes, carbonyls, aromatic
500 compounds and a number of other volatile organic species. The least volatile fraction has been
501 shown to range from 10 to 20% of the total organic emissions (Presto et al., 2011) and its presence
502 is particularly challenging, because it can react in the atmosphere and may undergo condensation in
503 the exhaust plumes leading to aerosol particles or volatile coating of pre-existing particles (Lee et
504 al., 2010; Miracolo et al., 2011). This latter component is named volatile PM, however there is
505 today a considerable controversy about its definition (Kinsey, 2009). Such particles may act as
506 condensation nuclei or may interact with soot to form condensation nuclei and thus may have
507 effects on cloud formation, precipitation and climate. In addition, additional compounds may
508 subsequently originate from the aging of exhausts following a chain of oxidation with atmospheric
509 oxidants and gases.

510

511 The relative amount of exhaust emissions depends upon combustor temperature and pressure, fuel
512 to air ratio and the extent to which fuel is atomised and mixed with inlet air (Anderson et al., 2006).
513 It is well recognised that the amounts of many pollutants may vary considerably with the engine
514 technology, model and especially with the thrust. For example Slemr et al. (1998, 2001) and Spicer
515 et al. (1992; 1994) reported that hydrocarbon emissions can be dependent upon engine type, use and
516 maintenance history as well as fuel composition.

517

518

519

520 **4.1 Geographical and Vertical Distributions of Flights**

521 Based upon the main air traffic routes, a series of studies have discussed the geographical and
522 vertical distributions of fuel consumption, which can be used to further assess the relative
523 emissions from aviation (e.g., Kim et al., 2007; Wilkerson et al., 2010; DeWitt et al., 2011; Olsen et
524 al., 2013; Simone et al., 2013). Due to the geographical distribution of civil aviation in the 2000s,
525 the global fuel burn by domestic flights is dominated by the North America and Caribbean regions,
526 while fuel consumed by international flights is dominated by Asia, North America and the
527 Caribbean, and Western Europe and North Atlantic (Kim et al., 2007). Using the Aviation
528 Emissions Inventory Code (AEIC, Stettler et al., 2011) Simone et al. (2013) estimated the fuel burn
529 by country of origin/destination in 2005 and reported that the USA was the most important (59.1
530 Tg), followed by Japan (9.7 Tg), UK (9.4 Tg), China (8.5 Tg, excluding Hong Kong), Germany (6.7
531 Tg) and France (5.4 Tg). A map showing the column sum of global fuel burn from scheduled civil
532 aviation in 2005 is provided in Figure 4a. Other studies have been carried out to estimate annual
533 fuel consumption and pollutant emissions more locally: for example Fan et al. (2012) assessed the
534 fuel consumption and emissions for each airline in China in 2010.

535

536 Kim et al. (2007) and Lee et al. (2007) used the System for assessing Aviation's Global Emissions
537 (SAGE) model to estimate the vertical profiles of commercial aviation and pointed out that the
538 highest fuel burn and emissions are between 9 and 12 km, which corresponds to typical cruise
539 altitude. Generally, most studies also reported that about 5–7% of total jet fuel is consumed within
540 1 km above ground level during airport operations (Kim et al., 2007; Simone et al., 2013), and
541 Olsen et al. (2013) reported a comparison of the annual global vertical distribution of fuel burn by
542 the commercial aviation deriving from different estimates (Figure 4b). Although most studies have
543 concluded that 5-10% of fuel is burned below 1000 m, aircraft operations within airports may
544 further increase fuel consumption due to the acceleration and deceleration of the engines following

545 airport congestion (Anderson et al., 2005; Nikoleris et al., 2011) or due the unaccounted use of fuel
546 for APUs (Ratliff et al., 2009).

547

548 **4.2 Emissions at Ground**

549 **4.2.1 *Landing and take-off (LTO) cycles***

550 The emissions of all aircraft engine must comply with applicable standards promulgated by the
551 International Civil Aviation Organization (ICAO, 2008) and measured upon the landing and take-
552 off (LTO) cycles. A LTO cycle refers to all the operations the aircraft carry out below 3000 ft above
553 field elevation (equivalent to 914 m) over a specific range of certifiable operating conditions and
554 includes four stages in terms of both engine thrust settings (expressed as a percentage of maximum
555 rated thrust, or F_{00}) and typical time in each specific mode of operation (time-in-mode, TIM). The
556 3000 ft height roughly corresponds to the atmospheric mixing height, i.e. the lower part of the
557 troposphere within which pollutants emitted at ground-level mix rapidly (e.g., Schäfer et al., 2006).
558 The LTO cycles are designed for aircraft engines manufactured after 1985 whose rated output is
559 greater than 26.7 kN and aim to guarantee they not exceed certain regulatory environmental limits
560 for a series of pollutants, namely unburned total hydrocarbons, carbon monoxide, nitrogen oxides
561 and smoke number (SN). This latter parameter is roughly representative of the amount of soot an
562 engine generates (e.g., Wayson et al., 2009; Stettler et al., 2013a,b). In the first LTO phase the
563 aircraft descends from cruising altitude toward the runway and lands at the airport. This phase is
564 named “approach” and is estimated as lasting for 4 min with engines at 30% F_{00} . After landing, the
565 aircraft enters in the “idle” phase which include all the ground-based operations: it proceeds at a
566 low speed to the gate (taxi-in), remains on stand-by for the loading and unloading operations and
567 again prepares for take-off proceeding towards the runway (taxi-out). Idle lasts 26 min and the
568 engines are required to be at 7% F_{00} . The subsequent operating modes include the “take-off” with
569 engines stressed to the full thrust (100% F_{00}) for 0.7 min, and the “climb” (85% F_{00} for 2.2 min) up
570 to 3000 ft height. A standardised LTO cycle is shown in Figure 5.

571 **4.2.2 *Engine ground running procedures***

572 In addition to the operations falling within LTO cycles, the ground running procedures (GRPs) may
573 lead to further emission loads from aircraft engines at airports. GRPs refer to the operation of some
574 or all engines carried out on the ground for the purpose of functionally checking the operation of
575 either engines or aircraft systems. GRPs are therefore an essential part of the operation of any
576 airliner prior to the release to service of an aircraft from maintenance. The main reasons for running
577 the engines on the ground are (Buttress and Morris, 2005): (i) check starts after minor maintenance
578 actions; (ii) runs at no more than ground idle to ensure that the engine operates correctly after
579 maintenance action, these include thrust reverser function checks, etc.; (iii) runs at powers greater
580 than ground idle to check the correct operation of certain valves, leak checks, etc. To date, only few
581 studies take into account the emissions from GRPs, but their importance for the atmospheric loads
582 of some pollutants cannot be neglected. For example, Buttress and Morris (2005) showed that GRPs
583 at London Heathrow airport release approximately $15.6 \text{ Mg y}^{-1} \text{ NO}_x$. Mazaheri et al. (2011)
584 investigated the annual emissions of particle number, particle mass and NO_x throughout the LTO
585 cycles and GRP at the Brisbane Airport and showed that annual emissions account for less than 3%.
586 Despite the evidence that GRPs may have a substantial impact on local air quality at airports, up to
587 now they have received only minor consideration. GRPs are not yet regulated internationally and
588 must comply only with local regulatory requirements imposing limitations on the locations, times
589 and engine thrust levels employed during ground running which may differ from one airport to
590 another.

591

592 **4.2.3 *Limitations in the use of standard LTO cycles***

593 The use of standard LTO cycles as a surrogate for typical aircraft operations close to the ground
594 represents an approximation and is not always representative of operations at airports. One
595 limitation is that the ICAO engine emissions standards are applied through national and multi-
596 national certification processes to turbojet and turbofan engines, but not turboprop, turboshaft and

piston engines (ICAO, 2011). This limitation may be negligible at large airports, where most traffic is due to common airliners equipped with TF engines, but may represent a major approximation for small and medium-sized airports where small, private, business and regional aircraft account for a large portion of flight traffic. In addition, despite LTO cycles having been designed to model optimally all the operational procedures of aircraft in the vicinity of airports, sometimes they are not well adapted to engine settings and actual TIM, which depend upon pilot' technique, fleets, airport layouts and flight traffic. In fact, default ICAO TIM are not representative of real operations and are for certification purposes. Consequently, although some inventories account for the deviations from the ICAO default TIMs and thrust settings, some deviations from the standardised LTO procedures may occur during actual LTO cycles. This inevitably leads to some differences between actual airport operations and emission inventories used in modelling studies. The main deviations/limitations are:

609

- reduced thrust during take-off. This practice is often carried out for performance and cost-efficiency reasons (ICAO, 2011) and has been widely observed on operational runways (Carslaw et al., 2008; Herndon et al., 2008); it may depend on aircraft weight and weather factors (Morris, 2002) and is often largely unknown (Carslaw et al., 2008). Since the emissions of some pollutants increase monotonically with the thrust (e.g., NO_x), this could lead to an overestimation of emissions from airports;
- lower thrust at idle/taxi mode. It has been reported that most aircraft use a thrust of 3%–4% F₀₀ instead of 7% (Morris, 2005a,b; Nikoleris et al., 2011 and reference therein) during idle operations. Since most pollutants emitted in exhaust plumes are strongly increased at decreased power settings (CO and generally all hydrocarbons), this may lead to underestimation of emissions at airports. In this context, Wood et al. (2008b) suggested that the thrust used in taxi operations can be split in two modes, i.e. 'ground idle' carried out at

4% F_{00} and ‘taxiway acceleration’ with thrust settings up to 17%. Moreover, higher thrust levels are sometimes used for turning;

- acceleration and deceleration of the engines or stop-and-go situations. This is mainly the result of congestion on taxiways and is known to be responsible for significant increases in fuel consumption and increased emissions (Anderson et al., 2005; Nikoleris et al., 2011). For example Morris (2005a) reported that instant accelerations up to 10% F_{00} and lasting ~10 s may occur at London Heathrow airport when aircraft cross an active runway or make a sharp turn. Due to this, the entire taxiway phase of operation using a uniform engine thrust level have been also recognised as problematic for emission inventory estimates because of the nonlinear emission rate of many compounds at low power (Herndon et al., 2009);

- use of a reverse thrust phase during landing. Reverse thrust is applied to assist mechanical brakes in slowing down the landing aircraft and is not generally required for normal operations onto a dry runway (ICAO, 2011). However, it generally occurs with idle thrust power as a prudent safety precaution, and under some circumstances it may also occur at power higher than 10% F_{00} (Morris and Easey, 2005; Stettler et al., 2011). Generally, reverse thrust is applied for 10–20 s (Fanning et al., 2007; Stettler et al., 2011), but may vary as a function of the landing velocity, runway length and aircraft weight;

- the evident differences between the standard TIM, which is used as part of the ICAO engine emissions certification processes, and the actual TIM used at airports (e.g., Unique, 2004; Watterson et al., 2004; Patterson et al., 2009; Stettler et al., 2011; Mazaheri et al., 2011; Khadilkar and Balakrishnan, 2012). For example, Patterson et al. (2009) and Khadilkar and Balakrishnan (2012) observed that total fuel burn during departures and arrivals at airports is generally overestimated by the ICAO method with respect to emissions computed from real-time aircraft flight data. Other studies have also reported measured TIM at airports: Unique (2004) reported TIM in Zurich airport and detected differences in all the LTO phases: idle (-43%), approach (+10%), climb (-77%) and take-off (+129%) which have been estimated to

have a strong impact on the calculation of emissions, resulting in reduced fuel flow (−38%) and NO_x emissions (−31%);

- the composition of the fleet that serves an airport and the weight of the aircraft. Since the ICAO certifies the engines and not the full aircraft, some airplane characteristics, mainly the aircraft weight, may have a key role in determining the emissions. Furthermore, in addition to the mass of the aircraft, its load of fuel, passengers and goods affect the overall weight: it is reported that passengers, crew and luggage usually add 6-15% to aircraft weight (Hu et al., 2009). Most of those factors vary from flight to flight, are largely unknown and may have direct implications for reduced thrust during take-off. In fact, it should be inferred that the increase of the aircraft weight has direct effects upon the thrust levels needed for carrying out usual LTO operations. For example, Carslaw et al. (2008) studied the NO_x emissions at London Heathrow and found evidence for statistically significant differences in the emissions from the same engine type used on the same aircraft frame. Among other factors, they speculated that the aircraft weight could be a cause. In a study conducted in eight major busy airports, Turgut and Rosen (2010) detected significant differences in the emissions of some pollutants and concluded that every airport has LTO cycles carried out by aircraft with different characteristics and, consequently, emissions. Another recent study by Turgut et al. (2013) showed a good relationship between aircraft mass and the NO_x emission during take-off and climb, which supports the concept of an explicit relationship between the aircraft weight and emissions. There is a general lack of knowledge about the relationships between aircraft mass and emissions, although some recent studies have indicated that heavier aircraft also emit more particles (Zhu et al., 2011).

Recent studies assessing airport emissions have proposed and used LTO cycles which are much more complex than those standardised by the ICAO. For example, in a study of the air quality and public health impacts of UK airports, Stettler et al. (2011) used specific TIMs derived from

Watterson et al. (2004) and Underwood et al. (2004) composed of 12 phases, namely approach, landing roll, reverse thrust, taxi-in, taxiway acceleration, APU, taxi-out, taxiway acceleration, hold, take-off, initial climb and climb-out. Proposed TIMs were developed by analysing the common procedures of an A320 aircraft at London Heathrow, but may vary by aircraft size category. Other studies (e.g., Ratliff et al., 2009), used models, such as the Emissions and Dispersion Modelling System (EDMS), which also requires jet fuel quality data, main engine and APU specifications, aircraft weight and ground operating time to generate more reliable emission estimates.

681

4.2.4 *The emission indices (EIs)*

The emissions during standardised LTO cycles are then reported as emission indices (EIs) expressed as mass of pollutant emitted per unit mass of fuel burned. Fuel-based emission indices for the compound X are calculated according to:

$$EI(X) = F_c \cdot (M_X / M_{CO_2}) \cdot (\Delta X / \Delta CO_2)$$

where F_c represents the stoichiometric calculation of CO_2 produced per kilogram of fuel consumed (with units $g\ CO_2\ kg\ Fuel^{-1}$) assuming complete combustion and given a particular hydrogen to carbon ratio (e.g., Herndon et al., 2004). M_X and M_{CO_2} are the molecular weights of the compound X and CO_2 , respectively, and ΔX and ΔCO_2 are the enhancements of compound X and CO_2 within the plume, respectively (e.g., Anderson et al., 2006). Unless specified differently, by convention $EI(NO_x)$ is defined in terms of NO_2 and therefore the mass of NO_x emissions is:

$$NO_x\ as\ NO_2 = NO_2\ emissions + NO\ emissions \cdot M(NO_2) / M(NO)$$

where $M(NO_2)$ and $M(NO)$ are the molecular weights of NO_2 and NO , respectively. In a similar way it should be specified that $EI(hydrocarbons)$ is often referenced to methane (Wahner et al., 1995). ICAO maintains a databank of engine certification data for commercial aviation reporting EIs for the four selected pollutants (EASA, 2013). Emissions of a pollutant X from an engine can be therefore calculated using three parameters: the first two are provided by the ICAO databank and are the main engine $EI(X)$ and the engine fuel flow, i.e., the burned fuel at a defined power setting

700 (expressed as kg s^{-1}); the third parameter is the time-in-mode (TIM), i.e. the time the engines spend
701 at an identified power setting (ICAO, 2011):

702
$$\text{Emission}(X) = \text{EI}(X) \cdot \text{TIM} \cdot \text{fuel flow}$$

703 Analogous to the EI for the emitted pollutant, emission indices for the number of particles have
704 been commonly reported in the literature. For convention, they are here reported as EI(#).

705 Using ICAO EIs and standardised LTO TIMs, Figure 6, 7 and 8 report a reprocessing of the data
706 included in the ICAO databank. In particular, Figure 6 shows the total burned fuel and the mass of
707 emitted pollutants (CO , NO_x and hydrocarbons) during a complete LTO cycle, i.e. the sum of
708 standardised time in each mode per fuel flow per average EI at each of the four power settings
709 (ICAO, 2013); data are organised to show the changes in the ICAO emission data for in-use engines
710 certified from 1973 to present (five year steps). Since different engines have different
711 characteristics, including the thrust force, Figure 6 also shows the ratios between the fuel burned
712 during complete LTO cycles and the engine maximum rated thrust (in kN) to normalise the fuel
713 consumption of the engine power. Figure 7 summarises the ICAO EI data (all in-use engines
714 certified from 1976 to today) per each LTO stage, expressed as g pollutant emitted per kg fuel
715 burned. Figure 8 shows the total burned fuel and emissions per each LTO phase, i.e. the product of
716 EIs per standardised time in each phase per fuel flow. The reprocessing of ICAO data does not take
717 into account the number of units produced for each engine model, but only the different models
718 produced and still in service in April 2013 (and included in the ICAO databank), regardless of
719 manufacturer, type and technology. Moreover, data refer to single engines, and generally
720 conventional aircraft are equipped with 1 to 4 engines. Therefore the sole purpose of the
721 reprocessing of ICAO data is to report qualitatively the trends in fuel consumption and emissions
722 for in-use TF engines.

723

724 Currently, the scientific literature includes several studies aiming to give EIs for comparison with
725 reported ICAO databank certification data and for many other components, including particulate

726 matter, elements, ions and speciated hydrocarbons. However, such data are often sparse and results
727 poorly comparable. Most studies were carried out using single or a few engine types, under certain
728 environmental conditions, without a standardised thrust and/or often using different measurement
729 techniques and instrumental set-up. Table 3 lists the most recent studies available in the literature
730 reporting EIs for various engines in aircraft and helicopters. The table also shows some information
731 (if available) about tested aircraft, engine models, selected thrust, type of fuel, sampling
732 methodologies and analytical techniques. Table 4 provides a list of recent studies which measured
733 EIs during real aircraft operations at airports. Most of the data in such studies (both engine tests and
734 real world operations) are summarised in the Supplemental Information Tables SI1, SI2, SI3 and
735 SI4, which provide detailed information about the EIs for many gaseous pollutants, speciated
736 hydrocarbons, particle number, particle mass (including soot) and species/ions in particulate matter,
737 respectively. Note that specific thrust levels provided in the tables are derived from the literature
738 and are categorised in five groups, named idle, approach, cruise, climb and take-off, on the basis of
739 the engine type. The thrust, expressed as F_{00} , is always provided along with the EIs. Additional
740 tested thrust levels (if available) are also reported, along with fuel and analytical methodologies.

741

742 **4.2.5** *Considerations about the EIs*

743 As indicated by the large number of studies in Tables 3 and 4, most of the literature provides results
744 through the calculation of EIs. When applied to the specific testing studies on engines or airplanes,
745 such methodology has the advantage of giving data easily comparable with EIs reported in the
746 ICAO databank. This may allow a better evaluation of the differences amongst tested engines and
747 technologies or, in case of the use of innovative analytical devices, allows a check the agreement
748 between data obtained and certified values. In contrast, expressing the results as EIs from studies
749 conducted during real-world operations at airports has both advantages and limitations. An
750 advantage of the specific studies may be comparison of the results with the ICAO data to detect
751 changes due to evolution of the exhaust plume, e.g. aging and gas-to-particle partitioning. Carslaw

et al. (2008) noticed that EIs do not give a clear indication of the absolute contribution of aircraft emissions to ground-level concentrations, which is important for assessing air quality at airports. Furthermore, they commented that the value of EIs may be substantially affected by limited knowledge of some important aircraft operational factors, such as the aircraft weight and thrust setting at take-off. A list of remaining studies conducted at airports and in their surroundings, which do not report data expressed as EIs, is provided in Table 5. In summary, Tables 3, 4 and 5 provide an overview of the most important studies reported in this review for the characterisation of aircraft emissions in both tests and real operations.

760

761 **4.3 Emissions at Cruise Altitudes**

Although injected at high altitudes, aircraft cruise emissions have been found to impact surface air quality through the mean meridional streamlines due to the polar, Ferrel, and Hadley cells (Barrett et al., 2010; 2012) and they are not currently regulated. Consequently, although this review focuses on airport emissions, a brief statement upon the aircraft emissions during cruise (8-12 km) is presented, as the majority of exhaust from aircraft is emitted at high altitudes (e.g., Gardner et al., 1997; FAA, 2005; Wilkerson et al. 2010; Whitt et al., 2011). A more exhaustive summary of the effects of both civil (subsonic) aviation in the upper troposphere and supersonic aircraft in the stratosphere is reported in two reviews by Lee and co-authors (Lee et al., 2009; 2010).

770

Impacts of aviation during cruising first focused the interest of the scientific community in the late 1960s in relation to contrail generation at high altitudes and the relative effect on climate (Reinking, 1968; Kuhn, 1970). Contrails are formed whenever the requisite conditions of either ice or water supersaturation exist within aircraft exhaust plumes (DeWitt and Hwang, 2005). Subsequently, in the early 1970s, concern grew over a possible role in stratospheric ozone depletion while interest in the impact of nitrogen oxide emissions on the formation of tropospheric ozone began in the late 1980s (Lee et al., 2009, and references therein). Subsequent studies (e.g., Wahner et al., 1995;

778 Brasseur et al., 1996; Schumann, 1997) investigated a number of emissions other than CO₂, and
 779 effects from aviation with potential effects on climate. To date there are a large number of studies
 780 characterising aircraft emissions during cruising (e.g., Fahey et al., 1995a,b; Busen and Schumann,
 781 1995; Schumann et al., 1996; Schlager et al., 1997; Paladino et al., 1998; Anderson et al., 1998a;
 782 Curtius et al., 1998; Brock et al., 2000; Schröder et al., 2000; Schumann et al., 2000; 2002; Curtius
 783 et al., 2002; Jurkat et al., 2011).
 784
 785 The RF of civil aviation emissions has been extensively studied (e.g., Prather et al., 1999; Wuebbles
 786 et al., 2007; Lee et al., 2009) and can be summarised in the following emitted compounds and
 787 processes, each having positive (+) or negative (–) forcing: H₂O (+); CO₂ (+); the atmospheric
 788 chemistry of NO_x causes the formation of tropospheric O₃ (+) but also the destruction of methane
 789 (–); oxidation of SO₂ results in sulphate particles (–); contrails (+); aviation-induced cloudiness
 790 (potentially +); soot, mainly composed of black carbon (+). Lee et al. (2009) estimated that
 791 aviation-induced RF in 2005 was ~55 mW m⁻², which accounted for 3.5% of global anthropogenic
 792 RF. In addition, black carbon emissions generated by aircraft at altitude have been shown to have a
 793 role in the formation of contrails (Schumann, 1996) and contrail-induced cirrus clouds, which affect
 794 the Earth's radiation balance by reflecting incoming solar radiation and by absorbing and re-
 795 emitting long wave radiation. The result is an additional positive RF of a magnitude similar to that
 796 of CO₂ (IPCC, 1999; Sausen et al. 2005; Lee et al., 2010). Recently, Azar and Johansson (2012)
 797 also assessed the non-CO₂ climate impact of aviation, including NO_x and contrails, and calculated
 798 the emissions weighting factors, i.e. the factor by which aviation CO₂ emissions should be
 799 multiplied to get the CO₂-equivalent emissions for annual fleet average conditions. Recently,
 800 Gettelman and Chen (2013) reported the climate impact of aviation aerosol. Although such studies
 801 highlighted the climate impact of aviation, it should be borne in mind that the magnitude of the total
 802 emissions of pollutants from aviation in terms of mass with direct and/or indirect effects on climate
 803 are one to two orders of magnitude smaller than from road transport or shipping (Balkanski et al.,

2010; Eyring et al., 2010). The study of aircraft emissions at cruise altitudes is very challenging mainly due to the obvious difficulty of sampling. Thus, measurements are commonly performed indirectly or extrapolated from data collected on the ground or in the laboratory. For this reason, the assessment of cruise emissions at altitude offers unique challenges to understanding the impacts of atmospheric emissions and their processing (Herndon et al., 2008, and reference therein). Computational models are available to extrapolate the test stand EI data to cruise altitude conditions (Baughcum et al., 1996b; Sutkus et al., 2001).

811

812 **4.4 Military Aircraft Emissions**

Despite most attention being given to civil aviation, a number of studies have also addressed emissions from military aircraft (e.g., Spicer et al., 1984; 1992; 1994; Heland and Schäfer, 1997;1998; Gerstle et al., 1999; 2002; Miller et al., 2003; Anderson et al., 2005; Brundish et al., 2007; Corporan et al., 2008; Cheng, 2009; Cowen et al., 2009; Spicer et al., 2009; Cheng et al., 2009; Cheng and Corporan, 2010). Despite the relatively high potential impact of military aircraft emissions under particular circumstances, the task of studying military emissions is very difficult. Unlike civil aviation, military operations generally do not work to set flight profiles and do not follow fixed plans (Wahner et al., 1995). In addition, national and military authorities are reluctant to disclose sensitive information either about operations or in-use technologies. The lack of comprehensive data about military operations makes realistic assessments of the contribution of military aircraft in terms of fuel consumption extremely difficult. In addition, some aircraft may have a dual function, such as the C-130 Hercules, which can be engaged in both military and civilian operations. Henderson et al. (1999) reported a historical breakdown of aviation fuel burn for civil and military aviation: in 1976 fuel burned by civil aviation was 64%, while military was 36%. In 1992 the percentages were 82% and 18%, respectively. Subsequent studies stated that military aviation fleets used 11% (19.5 Tg) of fuel in 2002 and estimated that the military contribution is in the range of 10-13% of total aviation emissions (Eyers et al., 2004; Waitz et al., 2005). Table 2

830 provides estimates of fuel consumption and exhaust emissions from military aviation by the
831 AERO2k model (Eyers et al., 2004). Among the large number of military aircraft, Cheng and
832 Corporan (2010) stated that the three classes of military engines T56, TF33, and T700/ T701C fitted
833 in the C130 Hercules, B-52 bomber and Apache/Blackhawk helicopters, respectively, consume
834 70%–80% of the USA military aviation fuel each year.

835

836 **4.5 Water Vapour**

837 Water is a key product of all hydrocarbon combustion and aircraft engines release H₂O as vapour
838 (Lewis et al., 1999). Water vapour is a greenhouse gas and its increase in the stratosphere (Solomon
839 et al., 2010) and the free troposphere (Sherwood et al., 2010) tend to warm the Earth's surface
840 (Prather et al., 1999). Water vapour, via latent heat released or absorbed during condensation and
841 evaporation cycles also play an active role in dynamic processes that shape the global circulation of
842 the atmosphere (Schneider et al., 2010). Moreover its effect on the formation of contrails and on the
843 enhanced cirrus generation in the upper troposphere can be relevant for additional global RF with
844 an indirect consequent potential increase of positive effects on global warming (Lee et al., 2009).
845 The annual and global-mean RF due to present-day aviation water vapour emissions has been found
846 to be 0.9 (range 0.3–1.4) mW m⁻² (Wilcox et al., 2012). The increased water vapour in the lower
847 troposphere may have secondary effects on precipitation, fog, visibility and some microphysical
848 processes.

849

850 An emission index of 1230±20 g H₂O kg Fuel⁻¹ is commonly reported for completely burnt fuel
851 (Lewis et al., 1999; Lee et al., 2010): this represents a little less than 30% of all combustion
852 products in aircraft exhaust (Figure 3). No differences in emission indices during idle, take-off and
853 cruise power settings are reported (Lewis et al., 1999), as emissions of H₂O are a simple function of
854 fuel consumption. The AERO2k inventories (Eyers et al., 2004) estimate a global emission of 217
855 Tg H₂O for 2002, 193 Tg from civil aviation and 24 Tg from military operations. Other more recent

estimates report 251 Tg H₂O in 2005 (Kim et al., 2007) and 233 Tg H₂O in 2006 (Wilkerson et al., 2010). However, the emissions of water by the global aircraft fleet into the troposphere are small if compared with fluxes within the natural hydrological cycle (IPCC, 1999) and thus water vapour from aircraft exhausts is not considered relevant for local air pollution and human health. An estimation of H₂O produced by aircraft below 1000 m can be assessed by considering the global use of fuel reported in the literature for LTO cycles: considering the total consumption of 13.9 Tg fuel in 2005 (Kim et al., 2007), a total emission of ~17 Tg H₂O can be estimated (Table 2). Considering the fuel burn breakdown provided by Simone et al. (2013) for the EU (3.1 Tg in 2005), a total of 3.8 Tg y⁻¹ H₂O are emitted within European countries.

865

866 **4.6 Carbon Dioxide**

Carbon dioxide is recognised as the main greenhouse gas, has a primary role in the Earth's climate warming and its behaviour within the atmosphere is simple and well understood (IPCC, 1999). Its main anthropogenic source is the combustion of fossil fuels: CO₂ emissions from fossil fuel combustion, including small contributions from cement production and gas flaring, were estimated to be 8.7±0.5 Pg C yr⁻¹ in 2008 an increase of 2% from 2007, 29% from 2000 and 41% from 1990 (Le Quéré et al., 2009). More recently, Peters et al. (2011) indicated that global CO₂ emissions from fossil-fuel combustion and cement production further grew by 5.9% in 2010, surpassing 9 Pg C yr⁻¹ principally due to the strong emissions growth in emerging economies. Once emitted, there are no important processes involving CO₂ formation or destruction and sinks occur principally at the Earth surface by exchange with the biosphere and the oceans (Solomon et al., 2007).

877

Carbon dioxide is the most abundant carbon-based effluent from aircraft engines (e.g., IPCC, 1999; Anderson et al., 2006; Lee et al., 2010) and Lewis et al. (1999) report that it accounts for ~72% of total combustion products (Figure 3). Typically, the EI(CO₂) from modern aircraft engines is 3160±60 g kg Fuel⁻¹ for complete combustion (Lewis et al., 1999; Lee et al., 2010) and emissions

882 of CO₂ are a simple function of fuel consumption (e.g., Owen et al., 2010). However, some studies
883 reported that EI(CO₂) decreases slightly at low thrust because incomplete combustion may result in
884 a relative increase of CO and hydrocarbons in the exhaust (e.g., Wey et al., 2006; Stettler et al.,
885 2011). The role of aviation in the rise of CO₂ emissions on a global scale may not be neglected and
886 a list of estimates of CO₂ emissions is provided in Table 2. In 1992, global aviation emissions of
887 CO₂ were about 2% of total anthropogenic sources and equivalent to about 13% of emissions from
888 all transportation sources (IPCC, 1999). The AERO2k inventories (Eyers et al., 2004) estimated a
889 global emission of 553 Tg CO₂ for 2002, 492 Tg from civil aviation and 61 Tg from military
890 operations, while a higher global emission of 733 Tg y⁻¹ was reported for 2005 (Lee et al., 2009),
891 accounting for approximately 3% of the total CO₂ emissions from the combustion of fossil fuels
892 (Howitt et al., 2011). Other estimates reported are 641 Tg CO₂ in 2005 (Kim et al., 2007) and 595
893 Tg CO₂ in 2006 (Wilkerson et al., 2010). As for H₂O, an estimate of CO₂ produced by aircraft
894 below 1000 m was derived by assuming a constant EI(CO₂) of 3160 g kg Fuel⁻¹ and by considering
895 the global use of fuel reported in the literature during LTO cycles in 2005 (Table 2). Results show a
896 global emission of 44 Tg CO₂ of which about 9.8 Tg y⁻¹ are emitted within Europe.

897

898 **4.7 Carbon Monoxide**

899 Carbon monoxide (CO) in the atmosphere is mainly generated by photochemical oxidation of
900 methane and nonmethane hydrocarbons as well as direct emissions from anthropogenic combustion
901 processes, such as vehicular exhaust, domestic heating, industrial emissions and biomass burning.
902 In the troposphere, CO has a chemical lifetime varying from 30 to 90 days and its major sink is
903 oxidation by hydroxyl radicals (Novelli et al., 1998; Seinfeld and Pandis, 2006). Its ability to form a
904 strong bond with haemoglobin to form carboxyhaemoglobin can cause adverse effects on human
905 health due to the reduction of blood oxygen-carrying capacity. At high exposure levels, CO can lead
906 to asphyxia, whereas at low doses it may cause impaired neuropsychological performance and risk

907 for myocardial ischemia and rhythm disturbances in persons with cardiovascular diseases (Samoli et
908 al., 2007; Bell et al., 2009).

909

910 Carbon monoxide is generally emitted in aircraft exhaust as result of incomplete combustion of jet
911 fuel. Emissions of CO are regulated by ICAO international standards and engine manufacturers
912 must provide emission indices for this pollutant during an LTO cycle (ICAO, 2008). In the last 40
913 years, the improvement of engine technology has led to a significant reduction in CO emissions
914 during the LTO cycle. Figure 6 shows a decrease in CO emissions at the end of the 1970s and
915 nowadays most newly certified engines emit less than 10 kg CO per complete LTO cycle.

916

917 Carbon monoxide emissions indices are highest at low power settings where combustor
918 temperatures and pressures are low and combustion is less efficient (Sutkus et al., 2001). Table SII
919 summarises values of EI(CO) certified by ICAO for specific in-use aircraft engines and also lists
920 EI(CO) for various military engines. Figure 7 reports the ICAO data (all in-use engines certified
921 from 1976 to today) as a function of LTO stages and shows that CO emission indices are generally
922 greater at lower thrusts. Generally, average EI(CO) for in-use commercial engines included in the
923 ICAO databank vary from 0.6 g kg Fuel⁻¹ at take-off power to 31 g kg Fuel⁻¹ at idle. Anderson et al.
924 (2006) observed large decreases in CO emissions with increasing engine power for various FSCs
925 (by a factor of ~8 from idle to 61% F₀₀) and reported that CO was observed to account for ~1% of
926 the total carbon emissions at engine idle, but emissions drop off at cruise thrust (61% F₀₀)
927 contributing <0.1%. Cain et al. (2013) measured emissions from a turbo-shaft engine burning
928 different types of fuel and observed a decrease of CO with increasing engine power mainly due to
929 improved combustion efficiency at higher power settings. Because of their predominant emission at
930 lower power settings, CO emissions from aircraft are of high relevance to air quality in the vicinity
931 of airports because of idle and taxi phases conducted at low thrust and which take up most of the
932 time aircraft spend at an airport. Figure 8 reports the total CO emissions for in-use engines during

933 the four LTO phases and shows that CO emissions during idle are generally two orders of
934 magnitude higher than climb and take-off phases.

935

936 After emission, CO may undergo to a series of chemical reactions in the troposphere involving
937 hydroxyl radical, O₂ and NO to form carbon dioxide, nitrogen dioxide, and ozone.

938

939 Some studies have derived EI(CO) directly from measurements during normal operation of idle and
940 taxi at airports and have revealed some considerable differences compared to ICAO data, with
941 results generally higher than those certified. For example, Heland and Schäfer (1998) reported an
942 EI(CO) of 51.8 ± 4.6 g kg Fuel⁻¹ at idle for a CFM56-3 engine, which was about 27-48% higher than
943 the ICAO data. Herndon et al. (2008) reported that EI(CO) observed in ground idle plumes was
944 greater (up to 100%) than predicted by engine certification data for the 7% thrust condition. Since
945 CO emissions increase with decreasing thrust, these studies seem to confirm that normal idle and
946 taxi operations at airports occur at lower thrust than the standardised ICAO LTO cycle, resulting in
947 more CO emitted than certified values (e.g., Schäfer et al., 2003).

948

949 Some studies have measured the carbon monoxide in ambient air at airports (e.g., Schürmann et al.,
950 2007; Heland and Schäfer, 1998; Yu et al., 2004; Herndon et al., 2008). In a study carried out at
951 two different airports, Yu et al. (2004) observed that aircraft are an important contributor to CO in
952 Hong Kong airport, whereas emissions from ground vehicles going in and out of the airport
953 dominated emissions at Los Angeles. A study carried out at Zurich airport (Schürmann et al., 2007)
954 demonstrated that CO concentrations in the vicinity of the terminals are highly dependent on
955 aircraft movements.

956

957

958

959 **4.8 Nitrogen Oxides and Nitrogen Acids**

960 Nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) in urban environments are principally emitted from fossil fuel
961 combustion as NO, as described by the extended Zeldovich mechanism (Lavoi et al., 1970):



965 NO plays an important role in atmospheric chemistry by rapidly reacting with ambient ozone or
966 radicals to form NO_2 on a timescale of minutes (Finlayson Pitts and Pitts, 2000; Seinfeld and
967 Pandis, 2006):



969 Other primary sources of NO_x in the troposphere are biomass burning, soil emissions, lightning,
970 transport from the stratosphere and ammonia oxidation (IPCC, 1999). NO_2 is a strong respiratory
971 irritant gas and its effects on human health have been extensively reviewed (Samoli et al., 2006;
972 Weinmayr et al., 2010; Chiusolo et al., 2011) indicating a relationship with cardiovascular and
973 respiratory diseases and mortality.

974

975 Nitrogen oxides are produced in the high temperature regions of the combustor primarily through
976 the thermal oxidation of atmospheric N_2 and therefore NO_x formation is sensitive to combustor
977 pressure, temperature, flow rate, and geometry (Sutkus et al., 2001). Additional NO_x may derive
978 from the combustion of the fuel-bound nitrogen: nitrogen in the fuel is not controlled or typically
979 measured, but it can range from near zero to perhaps 20 ppm (Chevron Corporation, 2006). Gardner
980 et al. (1997) estimated that 93% of NO_x from aircraft is emitted in the Northern Hemisphere and
981 ~60% at cruise altitudes. More recent estimates indicated that in 2005 the NO_x emitted during LTO
982 was 0.23 Tg (Kim et al., 2007), accounting for ~8% of global emissions from aviation.

983

984 NO_x is included in the parameters certified by ICAO. There is a difference in the molecular mass of
 985 NO and NO₂, and in the ICAO methodology data are reported as NO₂ equivalent (unless otherwise
 986 specified). Being sensitive to combustor pressure, NO_x emissions increase monotonically with
 987 engine thrust (Table SI1, Figure 7). Generally, EI(NO_x) for in-use engines included in the ICAO
 988 databank vary from 4±1 g NO_x kg⁻¹ burned Fuel⁻¹ at idle to 29±12 g NO_x kg⁻¹ burned Fuel⁻¹ at take-
 989 off power. However, despite the strong relationships to power settings, NO_x total emissions per
 990 each standardised LTO phase are pretty constant during idle, approach and take-off operations
 991 (Figure 8). Carslaw et al. (2008) measured individual plumes from aircraft departing Heathrow
 992 Airport and found that engines with higher reported NO_x emissions result in proportionately lower
 993 concentrations than engines with lower emissions. This result was hypothesised to be linked to
 994 aircraft operational factors, such as take-off weight and aircraft thrust setting, which therefore may
 995 have an important influence on concentrations of NO_x. Furthermore, Carslaw and co-authors
 996 reported that NO_x concentrations can differ by up to 41% for aircraft using the same airframe and
 997 engine type, while those due to the same engine type in different airframes can differ by 28%.
 998
 999 In recent years there has been a growing concern over emissions of primary NO₂ as a fraction of
 1000 NO_x from road traffic mainly because of the failure of NO_x emission reductions to deliver an
 1001 improvement in urban NO₂ concentrations (e.g., Jenkin, 2004; Carslaw and Beevers, 2004; Carslaw,
 1002 2005; Hueglin et al., 2006; Grice et al., 2009; Mavroidis and Chaloulakou, 2011; Cyrus et al.,
 1003 2012). The ratio of NO₂ to NO_x in aircraft emissions is diagnostic of combustor efficiency and
 1004 several studies reported that, unlike many other forms of combustion, the majority of the NO_x
 1005 emitted from modern high bypass TF engines at idle is in the form of NO₂. On the contrary, NO is
 1006 dominant at high power regimes. For example, Wormhoudt et al. (2007) performed ground
 1007 measurements and observed that emitted NO₂ may represent up to 80% of the total NO_x emissions
 1008 for a modern engine at low thrust and 7% at the highest power setting. Other studies (Timko et al.,
 1009 2010b,c; Wood et al., 2008b) reported that the NO₂/NO_x ratio may vary between 75% and 98% at

1010 low thrust, while for approach, thrust may range from 12% to 20%. Presto et al. (2011) observed
1011 that the NO/NO_x ratio increases from 0.2-0.3 at 4% F₀₀ to 1 at 30% and 85% F₀₀. Other
1012 measurements carried out within 350 m of a taxiway and 550 m of a runway during common
1013 airport operations indicated that 28–35% of NO_x exists in the form of NO₂ (Herndon et al., 2004).
1014 However it was reported that the relative abundance of NO and NO₂ are subject to large
1015 uncertainties due to conversion in the plumes and the contribution of other sources. The results of a
1016 study performed by Schäfer et al. (2003) using remote sensing methodologies suggested that NO
1017 was rapidly converted to NO₂ in the exhaust plume. The NO₂ formation and destruction processes
1018 of aircraft exhausts were investigated by Wood et al. (2008b), who observed that the NO₂/NO_x
1019 fraction is significantly higher in advected measurements than in engine tests. The results suggested
1020 that a significant portion of the NO in the exhaust can be converted into NO₂ by mechanisms that do
1021 not involve ozone.

1022

1023 Nitrogen oxides may also be oxidised to other reactive nitrogen species and the complete family of
1024 reactive nitrogen species is denoted as reactive odd nitrogen (NO_y), which includes the sum of NO_x
1025 and its oxidation products (HNO₃, HONO, NO₃[·], N₂O₅, HNO₄, peroxyacyl nitrates, alkyl nitrates
1026 and others). Nitric acid is the major oxidation product and increasing atmospheric concentrations of
1027 NO_x favour nitric acid formation as a result of the daytime gas phase recombination reaction of
1028 hydroxyl radical with NO₂. NO_x plays a key role in secondary inorganic aerosol formation
1029 (Finlayson-Pitts and Pitts, 2000; Seinfeld and Pandis, 2006).

1030

1031 High levels of NO_x, particularly NO₂, are a matter of concern for air quality near major airports. For
1032 example, current NO₂ concentrations breach the UK annual mean air quality objective (40 µg m⁻³)
1033 at some locations around Heathrow, London (UK) (UK Department of Transport, 2006; UK
1034 Statutory Instrument, 2007; HAL, 2011), while some exceedences of the Swiss annual mean NO₂
1035 limit value (30 µg m⁻³) have been observed near Zürich airport (Fleuti and Hofmann, 2005).

1036 However, as most airports are located in the vicinity of large cities, the contribution of airport-
1037 related emissions to those exceedences is hard to quantify due to the major influence of other
1038 sources, such as traffic and industry. For example, Yu et al. (2004) observed that ground vehicles
1039 were the dominant source of NO_x emissions at Los Angeles airport.

1040

1041 Although various studies have attempted to estimate the contribution of airport operations to
1042 ambient NO_x levels, the results are often conflicting. For example, Carslaw et al. (2006) estimated
1043 that Heathrow operations accounted for ~27% of the annual mean NO_x and NO₂ at the airfield
1044 boundary and less than 15% (<10 µg m⁻³) at background locations 2-3 km downwind of the airport,
1045 while Fleuti and Hofmann (2005) estimated the Zürich airport influence upon NO₂ to be below 1 µg
1046 m⁻³ at a distance of three or more kilometers. In both case studies concentrations of NO_x close to
1047 the airport were dominated by road traffic sources. A detailed emission inventory of UK airports
1048 was computed by Stettler et al. (2011), who pointed out that LTO emissions at London Heathrow in
1049 2005 accounted for about 8.19x10⁶ kg NO_x, of which more than 80% is in the form of NO. An
1050 emission inventory study of NO_x emissions at Zurich airport in 2003 (Unique, 2004) reported that
1051 most nitrogen oxides were released from LTO operations, while minor contributions were
1052 calculated for landside traffic, handling/airside traffic and airport infrastructure.

1053

1054 **4.8.1 Nitrous oxide**

1055 Apart from NO_x, other nitrogen species have been detected and analysed in aircraft exhaust plumes
1056 and at airports. Few data are available for the emissions of nitrous oxide (N₂O) and some are
1057 contradictory. Wiesen et al.(1994) examined nitrous oxide emissions from different commercial jet
1058 engines using different fuels and reported average EI(N₂O) ranging from 97 to 122 mg kg Fuel⁻¹.
1059 Heland and Schäfer (1998) further analysed N₂O using FTIR techniques and observed that N₂O
1060 emitted by a CFM56-family engine was under the detection limits at idle thrust and detectable at
1061 higher power settings, with a related EI(N₂O) of 1300 mg kg Fuel⁻¹. Conversely, Santoni et al.

(2011) measured N_2O emissions from a CFM56-2C1 engine and concluded that at low thrust EI N_2O were $110 \pm 50 \text{ mg kg Fuel}^{-1}$ (mean \pm standard deviation), while a drop of emissions was observed at higher thrust levels ($32 \pm 18 \text{ mg kg Fuel}^{-1}$).

1065

1066 **4.8.2 Nitrous acid**

1067 HONO is generated in the gas turbines via reaction of hydroxyl radical with NO (Wormhoudt et al.,
1068 2007; Brundish et al., 2007) and $\sim 1.1\%$ of the total NO_y is in the form of HONO by the engine exit
1069 (Lukachko et al., 1998). Anderson et al. (2005) measured nitrous acid (HONO) in the exhaust of a
1070 B757 and observed a clear power dependence, increasing with increasing power; at high power,
1071 over 2 ppmv of HONO was detected. The same authors (Wormhoudt et al., 2007) further reported
1072 an increasing EI(HONO) at increasing thrust, but also reported that the EI(HONO)/EI(NO_2) ratio
1073 decreases with increasing engine regimes. They found that HONO is a minor constituent (up to 7%)
1074 compared with NO_x . Herndon et al. (2006) measured NO_y at Logan airport in Boston (USA) and
1075 reported that the emission index for a B737 increased from idle ($2 \pm 1.9 \text{ g}(\text{NO}_y) \text{ kg Fuel}^{-1}$) to take-off
1076 ($19.5 \pm 3.9 \text{ g}(\text{NO}_y) \text{ kg Fuel}^{-1}$). Wood et al. (2008b) reported that HONO accounts for 0.5% to 7% of
1077 NO_y emissions from aircraft exhaust depending on thrust and engine type: 2–7% for low thrust and
1078 0.5–1% for high thrust (65–100% F_{00}). In conclusion, using data available in the literature, Lee et
1079 al. (2010) proposed that EI(HONO) should range between 0.08 and $0.8 \text{ g kg Fuel}^{-1}$. More recently,
1080 Lee et al. (2011) performed measurements of HONO from a DC-8 aircraft equipped with CFM56-
1081 series engines using both traditional and synthetic fuels and observed that the EI(HONO) increases
1082 approximately 6-fold from idle to take-off conditions, but plateaus between 65 and 100% of
1083 maximum rated engine thrust. This study also discussed the kinetics behind the HONO
1084 formation/destruction.

1085

1086 Jurkat et al. (2011) measured the gaseous nitrogen emissions in young aircraft exhaust plumes
1087 emitted by 8 different types of modern jet airliners in flight and calculated molar ratios of

1088 HONO/NO and HONO/NO_y of 0.038±0.010 and 0.027 ± 0.005, respectively. The relative
1089 EI(HONO) at cruise thrust was reported to be 0.31±0.12 g NO₂ kg Fuel⁻¹.

1090

1091

1092 **4.8.3 Nitric acid**

1093 Most studies of HNO₃ emissions were performed using experimental measurements with chemical
1094 ionisation mass spectrometry (CIMS) in both exhaust plumes at cruising altitudes (e.g., Arnold et
1095 al., 1992;1998a; Tremmel et al., 1998; Miller et al., 2003) and simulated gas turbines (Katrakou et
1096 al., 2004) or using plume models (e.g., Garnier et al., 1997; Kraabøl et al., 2002). Generation of
1097 HNO₃ is generally lower than HONO: Lukachko et al. (1998) reported that only ~0.07% of the total
1098 NO_y is oxidised to HNO₃ by the engine exit, while Lee et al. (2010, and references therein) reported
1099 EI(HNO₃) of 0.003–0.3 g kg Fuel⁻¹. Because of the very low levels expected in aircraft exhaust, few
1100 studies have been carried out on the ground. There is consequently a lack of data about nitric acid
1101 measured in engine exhaust plumes during real working conditions.

1102

1103 **4.9 Sulfur Oxides and Sulfuric Acid**

1104 **4.9.1 Sulfur oxides**

1105 Sulfur dioxide (SO₂) is emitted into the atmosphere from both natural (volcanic activity, grassland
1106 and forest fires) and anthropogenic sources, including crude oil and coal transformation processes,
1107 fossil fuel combustion, metal smelting and various industrial processes (e.g., Seinfeld and Pandis,
1108 2006; Smith et al., 2011). Exposure is associated with increased mortality and morbidity
1109 (Katsouyanni et al., 1997; Sunyer et al., 2003a) including cardiovascular admissions, particularly
1110 for ischemic heart disease (Sunyer et al., 2003b). Oxidation of SO₂ (S(IV)) is recognised as the
1111 major channel for the formation of atmospheric sulfuric acid (S(VI)), and sulfur trioxide (SO₃) is an
1112 important intermediate in the oxidation processes (Vahedpour et al., 2011). Consequently, SO₂ has
1113 an indirect effect on acid deposition and a key role in the aerosol system by acting as sulphate

precursor. Since sulphate aerosol is known to modify the direct and indirect RF, SO₂ also has an indirect influence on climate.

Sulfur dioxide is the overwhelmingly predominant S-containing species in aircraft exhaust (Anderson et al., 2005; Lee et al., 2010) and originates mainly from the oxidation of fuel sulfur in the engines (Brown et al., 1996a; Schumann et al., 2002). Therefore, SO₂ emissions may vary greatly as a function of FSC. In the past, studies were carried out to analyse and model the sulfur emissions of aircraft and to estimate their role in the formation of visible contrails (e.g., Busen and Schumann, 1995; Schumann et al., 1996; Brown et al., 1996b; 1997; Arnold et al., 1998a).

Generally an emission index of 0.8–1.3 g of SO_x (as SO₂) per kg Fuel was reported for complete combustion (e.g., Lewis et al., 1999; Kim et al., 2007; Lee et al., 2010; Presto et al., 2011), however measurements at flight altitudes have showed that sulfur dioxide varies with the average FSC (e.g., Arnold et al., 1998a; Schumann et al., 1998). For example, Hunton et al. (2000) reported that the EI(SO₂) varied from 2.49 g SO₂ kg fuel⁻¹ for a high-sulfur fuel (~1150 ppm S) in a test chamber to less than 0.01 g SO₂ kg fuel⁻¹ for a low-sulfur fuel (~10 ppm S). They also reported that there is no dependence of emission indices upon engine power.

In this context, it is very important to stress that no S is created or destroyed from the fuel to the exhausts, therefore for every fuel S atom there is a molecule of SO₂ or SO₃ at the exhaust plane (the SO₃ quickly converts to H₂SO₄). In this way the emission indices of total emitted S may vary according to the FSC, whereas the only uncertainties are in the speciation between S(IV) to S(VI) species, i.e. in the conversion efficiency, which is discussed fully later.

The importance of SO₂ emissions at local scale, i.e. near the airports, was highlighted by Yu et al. (2004), who found that sulfur dioxide was a good tracer of aircraft emissions at both Los Angeles and Hong Kong airports. However, on a global scale the aviation source is considered to be

secondary with respect to other major sources of SO₂: Kjellström et al. (1999) used a atmospheric general circulation model including the atmospheric sulfur cycle to investigate the impact of aircraft sulfur emissions on the global sulfur budget of the atmosphere and concluded that aviation accounted for about 1% of the total sulphate mass north of 40°N, where aircraft emissions are largest. In 2004, about 0.18 Tg of SO₂ was estimated to be emitted from aviation (Lee et al., 2010) using an EI(SO₂) of 0.8 g Fuel⁻¹. An estimation of SO₂ produced by aircraft below 1000 m can be computed by applying a constant EI(SO₂) of 0.8 g kg Fuel⁻¹ and by considering the global use of fuel reported by the literature during LTO cycles in 2005 (Table 2). Results show a global emission of 11 Mg SO₂ of which about 2.5 Mg y⁻¹ are emitted within Europe.

1149

1150 **4.9.2 Conversion of S(IV) to S(VI)**

1151 Despite SO₂ being the dominant S-species in aircraft exhaust emissions, a fraction can be further
 1152 oxidised to form S(VI) as SO₃ and H₂SO₄ (Lee et al., 2010). The presence of SO₃ has been
 1153 established in gas turbine engine exhaust and as attributed mainly to the oxidation of SO₂ by O
 1154 atoms (Arnold et al., 1998a) or by hydroxyl radicals in exhaust plumes (Tremmel and Schumann,
 1155 1999). The further reaction with water vapour rapidly converts SO₃ to sulfuric acid, according to
 1156 Stockwell and Calvert (1983); Stockwell (1994); Brown et al., (1996a) and Seinfeld and Pandis,
 1157 (2006):



1161 Starik et al. (2002) computed that ~1% of the sulfur is converted into SO₃ within the combustor and
 1162 about 10% into SO₃ and H₂SO₄ before the engine exit. Past numerical simulations of H₂SO₄
 1163 formation from atomic oxygen and hydroxyl radical in aircraft engines indicated that between 2%
 1164 and 10% of the fuel sulfur is emitted as S(VI) (Brown et al., 1996a; Lukachko et al., 1998).
 1165 However, current understanding indicates a more realistic value of 2% (or possibly less). These

1166 studies also indicate that S(VI) conversion in the turbine is kinetically limited by the level of atomic
1167 oxygen, resulting in a higher oxidation efficiency at lower FSCs. Katragkou et al. (2004) report that
1168 the limiting factor of this series of reactions is the oxidation of SO₂ by the hydroxyl radical, which
1169 is somewhat uncertain at the high temperatures in gas turbine engines. The knowledge of the
1170 mechanisms involving sulfur species and their interactions with H, O atoms and radicals occurring
1171 within a combustor is far from complete and are the subject of discussion (e.g., Blitz et al., 2003;
1172 Somnitz et al., 2005; DeWitt and Hwang, 2005; Yilmaz et al., 2006; Hindiyarti et al., 2007;
1173 Rasmussen et al., 2007; Wheeler and Schaefer, 2009; Hwang et al., 2010).

1174

1175 Once emitted, the gaseous sulfuric acid may act as an important precursor for aerosol because of its
1176 low vapour pressure. An understanding of the processes controlling sulphate aerosols is therefore
1177 essential to the study of the mechanisms of formation of particles generated by aircraft (e.g., Starik
1178 et al., 2004). For example, Arnold et al (1998a) reported no detectable levels of sulfuric acid in the
1179 gas phase behind an in-flight commercial aircraft, leading to the inference that initially formed
1180 H₂SO₄ experiences a rapid gas-to-particle conversion at plume ages <1.6 s. Sulfuric acid was
1181 measured in several other studies at cruising altitudes and for different FSCs (e.g., Fahey et al.,
1182 1995b; Busen and Schumann, 1995; Schumann et al., 1996; Curtius et al., 1998; Arnold et al.,
1183 1998a; Schröder et al., 2000; Schumann et al., 2000; Curtius et al., 2002) as well as in fuel
1184 combustion experiments at ground-level (Frenzel and Arnold, 1994; Curtius et al., 1998; 2002;
1185 Kiendler and Arnold, 2002; Sorokin et al., 2004) and during combustor testing (Katragkou et al.,
1186 2004). Curtius et al. (2002) reported H₂SO₄ concentrations measured in the plume were up to 600
1187 pptv for a 56 ppm FSC, while the average concentration of H₂SO₄ measured in the ambient
1188 atmosphere outside the aircraft plume was 88 pptv and the maximum ambient atmospheric
1189 concentration 300 pptv.

1190

1191 The abundance ratio, sometime named conversion factor ($\epsilon = (\text{SO}_3 + \text{H}_2\text{SO}_4) / \text{total sulfur}$) has been
1192 widely used to assess the ratio of S(VI) to total sulfur at the exit of engines. The literature offers
1193 numerous estimates or measures of ϵ . However, the results are often difficult to compare as they
1194 are derived by different methods, ranging from direct measurements, indirect computations and
1195 models. In addition, most studies take in account only particulate sulphate, while only a few studies
1196 have measured both particulate and gaseous phases. Anyway, Timko et al. (2010b) demonstrated
1197 that the conversion of S(IV) to S(VI) is independent of engine technology for most modern in-use
1198 engines. Earlier values of ϵ are well summarised in DeWitt and Hwang (2005), while most recent
1199 measurements and modelling studies of aircraft plume chemistry reported other direct, indirect and
1200 inferred values of ϵ . Generally, ϵ values between 1 and 3% are commonly reported. For example, ϵ
1201 values between 6 and 31% have been calculated for a B757 aircraft (Miake-Lye et al., 1998), while
1202 Schumann et al. (2002) observed ϵ between 0.34 and 4.5% for an old engine (Mk501) and $3.3 \pm 1.8\%$
1203 for a modern engine (CFM56-3B1). For low FSC, they also reported that ϵ was considerably
1204 smaller than implied by the volume of volatile particles in the exhaust, while for $\text{FSC} \geq 100$ ppm,
1205 sulfuric acid is the most important precursor of volatile aerosols formed in aircraft exhaust plumes
1206 of modern engines. Kiendler and Arnold (2002) inferred an ϵ value of $2 \pm 0.8\%$ for a M45H engine
1207 on the ground, while Curtius et al. (1998; 2002) reported $3.3 \pm 1.8\%$ in the plume of a B737-300
1208 aircraft in flight by measuring the total H_2SO_4 content in both gaseous and aerosol phases. The
1209 sulfur conversion fraction of an RB211 engine was computed by Starik et al. (2002) using a model
1210 and results showed that increases in FSC cause a minor reduction in ϵ , reporting values $\approx 9\%$, and
1211 $\approx 8.4\%$ for FSC of 0.04% and 0.3%, respectively. Wilson et al. (2004) and Sorokin et al. (2004)
1212 observed ϵ of $2.3 \pm 1.2\%$ in an A310 equipped with a CF6-series engine at an exhaust age of about 5
1213 ms from the combustor exit, while Jurkat et al (2011) derived ϵ for various in-flight aircraft and
1214 reported an average value of $2.2 \pm 0.5\%$, varying from a minimum of 1.2% for a Trent-series and a
1215 maximum of 2.8% for a CMF56-series engines. Wong et al. (2008) modelled the microphysical
1216 processes involved and suggested conversion efficiency of 1–2%. Timko et al. (2010b) reported ϵ

1217 ranging from 0.08% to 0.01%, while Kinsey et al. (2011) suggest a median value of 2.4%. Petzold
 1218 et al. (2005b) reported that sulfur partitioning at 150°C was 97 % $\text{SO}_2 \leq 2.7\%$ gaseous $\text{H}_2\text{SO}_4 <$
 1219 0.3% chemisorbed H_2SO_4 at soot particle surface. Regarding the relative abundance of the two
 1220 S(VI) species, during the COMS experiments Sorokin et al. (2004) reported that SO_3 represented
 1221 the major fraction of S(VI) in the exhaust behind the combustor and that SO_3 conversion to H_2SO_4
 1222 takes place in the sampling line where the exhaust gases spend a sufficiently long time and where
 1223 the temperature is markedly lower than in the hot exhaust. Other experimental measurements made
 1224 during the EXCAVATE experiment by Anderson et al. (2005) led to the conclusion that the fraction
 1225 of total sulfur that existed as SO_3 would have to be less than 0.005%.

1226

1227 According to the conversion factors for sulfur species and taking in account the mass conservation
 1228 of S in the exhaust plumes (no S is created or destroyed from the fuel to the exhausts), the
 1229 computation of the EIs can be assessed by applying:

$$1230 \quad \text{EI}(\text{SO}_2) = (\text{M}(\text{SO}_2)/\text{M}(\text{S})) \cdot \text{FSC} \cdot (1-\varepsilon)$$

1231 and

$$1232 \quad \text{EI}(\text{SO}_4^{2-}) = (\text{M}(\text{SO}_4^{2-})/\text{M}(\text{S})) \cdot \text{FSC} \cdot \varepsilon$$

1233 where M() represents the molecular weights of sulfur species, FSC is the fuel sulfur content and ε
 1234 is the S(IV) to S(VI) conversion efficiency as a fraction, e.g. 0.02 and a unit conversion may be
 1235 necessary (e.g. if FSC is in expressed ppm, etc).

1236

1237 Another important consideration concerning the sulphate derived from aircraft engines was pointed
 1238 out during the APEX-1 project, which was primarily developed to investigate the effects of fuel
 1239 composition on emissions at various power settings (e.g., Wey et al., 2006; Knighton et al., 2007;
 1240 Yelvington et al., 2007; Onash et al., 2009). General results from the testing of a CFM56-series
 1241 engine showed a strong linear relationship ($r^2=0.93$) between FSC and emission indices for

1242 sulphate, which can be approximately described by the linear equation $EI(\text{sulfur in mg kg}$
1243 $\text{Fuel}^{-1}) = 0.0136 \cdot \text{FSC}(\text{in ppm}) + 4.4952$ (Kinsey, 2009).

1244

1245 **4.10 Ozone**

1246 Ozone (O_3) is a reactive oxidant gas playing a key role in photochemical air pollution and in
1247 atmospheric oxidation processes. Ozone is associated with decrements in respiratory function and
1248 death from respiratory causes (Jerrett et al., 2009; Yang and Omaye, 2009). Although in the upper
1249 atmosphere it acts as a barrier for ultraviolet radiation, in the lower troposphere is a secondary air
1250 pollutant generated through a series of complex photochemical reactions involving reactive
1251 hydrocarbons, solar radiation and NO_2 (Finlayson-Pitts and Pitts, 2000; Seinfeld and Pandis, 2006).

1252

1253 Ozone is not primarily produced by aircraft engines, however some ozone precursor such as CO,
1254 NO_x and VOCs are emitted from the exhaust and may subsequently increase the boundary layer O_3
1255 pollution. Note that, amongst the ozone precursors, both CO and many VOCs are mainly emitted at
1256 low power settings during airport taxi and idle operations, while NO_x is mainly released during
1257 take-off and climb phases, when engines reach higher thrusts. It is reported that NO emissions,
1258 which are dominant at highest thrusts, initially cause local ozone reductions in aircraft plumes
1259 (Kraabøl et al., 2000a,b) following:



1261 but subsequently the photolysis of NO_2 may form atomic oxygen which reacts with molecular O_2 to
1262 form O_3 :



1265 where M is N_2 , O_2 or another molecule absorbing the excess energy to stabilise the ozone formed
1266 (Seinfeld and Pandis, 2006). A contrary effect, i.e. a decrease in O_3 concentrations, may also occur
1267 due to the reaction of ozone with other compounds emitted from aircraft. For example, it is

1268 recognised that alkenes, which are emitted in the exhaust plumes, are susceptible to reaction with
1269 ozone forming primary carbonyls and bi-radicals (e.g., Grosjean et al., 1994; Seinfeld and Pandis,
1270 2006) and consuming O₃.

1271

1272 Although the effects of aircraft emissions on ozone depletion in the upper troposphere and
1273 stratosphere have been addressed by IPCC (1999) and the European 6th Framework 'ATTICA'
1274 (Assessment of Transport Impacts on Climate Change and Ozone Depletion) project (Lee et al.,
1275 2010), less attention has been given to the effects within the boundary layer due to emissions during
1276 LTO operations.

1277

1278 **4.11 Hydrocarbons**

1279 Unburned hydrocarbons (UHC) are emitted as a result of the inefficiency of jet turbine engines to
1280 completely convert fuel to CO₂ and H₂O (Knighton et al., 2009). Although the levels of UHC
1281 emitted by aviation are considered negligible relative to emissions from surface transportation
1282 systems such road traffic, they may cause adverse health effects on exposed people, including
1283 workers and travellers at airports, and residents who live near large hubs. Therefore, UHC are
1284 included as parameter to be monitored during the LTO cycles by ICAO (ICAO, 2008). Analyzing
1285 the data provided by the ICAO databank (EASA, 2013), a large range in the magnitude of UHC
1286 emissions between different engine models can be observed. Moreover, ICAO data clearly show
1287 that the emission of UHC during complete LTO cycles have fallen considerably since the 1970s
1288 (Figure 6), mainly due to the development of more efficient technologies.

1289 Unfortunately, the UHC parameter used by ICAO only refers to the lump sum of all hydrocarbons,
1290 including contributions from methane, and no corrections are made for background levels within the
1291 engine intake air (Anderson et al., 2006; Lee et al., 2010). Consequently, UHC data give no
1292 information on the large number of specific non-methane hydrocarbons (NMHCs) nowadays
1293 identified, and in some cases quantified, in aircraft exhaust plumes (Wilson et al., 2004; Anderson

1294 et al., 2006; Lobo et al., 2007; Agrawal et al., 2008; Herndon et al., 2009). This fact clearly
1295 represents a significant gap in the knowledge of impacts of aircraft on both environmental and
1296 human health endpoints, because of the very different physicochemical and toxicological properties
1297 of each class of organic compounds. Most emitted VOC are known ozone precursors, many are
1298 particle precursors and can impact visibility after particle formation. Some compounds are known
1299 or are suspected to have adverse effects on human health and the environment. Among the
1300 hydrocarbons emitted in aircraft exhaust, 14 species (12 compounds and two groups of complex
1301 organic compounds) are present in the Hazardous Air Pollutants (HAP) list compiled by the
1302 USEPA (Federal Aviation Administration, 2003). These compounds are 1,3-butadiene, *n*-hexane,
1303 acetaldehyde, xylene, acrolein, propionaldehyde, benzene, styrene, ethylbenzene, toluene,
1304 formaldehyde, lead compounds and polycyclic organic matter as 7 and 16 PAH groups.

1305

1306 In the last 20 years, various research programmes and experiments have been carried out to give
1307 more detailed data on the speciated hydrocarbon emissions of aircraft engines. Among others, some
1308 milestones are listed hereafter. Spicer et al. (1984;1994) measured detailed organic emissions for
1309 the CFM56- class engines burning various JP-grade fuels; Gerstle et al. (1999; 2002) reported UHC
1310 emission rates for several military engines not included in the ICAO databank; the EXCAVATE
1311 campaign (Anderson et al., 2005; 2006) investigated the speciated-hydrocarbon emissions from an
1312 RB211-535-E4 engine at two different fuel sulfur levels; Herndon et al. (2006) investigated a set of
1313 hydrocarbons from in-use aircraft at Boston Logan International Airport; the APEX-1 campaign
1314 (Wey et al., 2006) reported the hydrocarbon speciation for a CFM56-2C1 engine using fuels with
1315 differing FSC (Knighton et al., 2007; Yelvington et al., 2007); Schürmann et al. (2007) sampled
1316 volatile organic compounds in diluted exhausts; the JETS/APEX-2 and APEX-3 campaigns (Lobo
1317 et al., 2007; Kinsey, 2009) reported data for speciated hydrocarbons in both a staged aircraft test
1318 (Yelvington et al., 2007; Wey et al., 2007; Agrawal et al., 2008; Timko et al., 2010c) and at airports
1319 (Wood et al., 2008b; Herndon et al., 2009); Knighton et al. (2009) consolidated earlier data from

1320 Spicer et al. (1984;1994), EXCAVATE and APEX studies; Cain et al. (2013) measured speciated
1321 hydrocarbon emissions from a TS engine burning various (conventional, alternative and surrogate)
1322 fuels.

1323

1324 Although those studies have yielded much useful information for characterizing the emissions of
1325 hydrocarbons, to date there is still a great deal of work to be done, many chemical and physical
1326 characteristics remain unclear, and some conflicting results need to be further investigated. Firstly,
1327 Spicer et al. (1984) reported that a significant percentage (30%–40%) of the total hydrocarbon
1328 emissions at idle are made up of a large number of exhaust compounds with aliphatic,
1329 cycloaliphatic and aromatic structures, predominantly ethylene, propylene, acetylene, 1-butene,
1330 methane, and formaldehyde. This latter carbonyl was found to be the predominant aldehyde present
1331 in the exhaust. In addition to byproducts of combustion, some studies (Spicer et al., 1992;1994;
1332 Slemr et al., 2001) also observed that unburned/unreacted fuel compounds are emitted in the engine
1333 exhaust from fuel cracking and incomplete combustion. Spicer et al. (1984) reported that
1334 compounds from unburned fuel may represent a major component of exhausts and that they are
1335 mainly composed of normal C₁₀–C₁₆ paraffins with smaller amounts of alkyl substituted aromatics,
1336 cycloparaffins, and branched alkanes. The unburned fuel component was also observed to be
1337 virtually eliminated at the 30% and 80% F₀₀ conditions, when concentrations of all of the individual
1338 hydrocarbons are very low. Similar results were reported by Slemr et al. (2001) in both modern
1339 commercial high bypass TF engines (CFM56-2C1) and older technology engines (Rolls Royce
1340 M45H Mk501) with emissions dominated by alkenes and alkynes due to fuel cracking and aromatic
1341 compounds arising from unburned fuel.

1342

1343 These pioneering results were largely confirmed by more recent studies, which generally reported
1344 that emitted hydrocarbons are composed of relatively light weight (C₂–C₆) species, including
1345 alkanes and alkenes, formaldehyde, methanol, ethylene, acetaldehyde, acetic acid, benzene, toluene,

1346 phenol, styrene, naphthalene and methylnaphthalenes (Slemr et al., 2001; Anderson et al., 2006;
1347 Knighton et al., 2007; Yelvington et al., 2007; Schürmann et al., 2007; Kinsey, 2009). The results
1348 of the whole APEX study (Kinsey, 2009) partially confirmed previous data, indicating that
1349 generally the gaseous hydrocarbon emissions of various engines primarily consist of formaldehyde
1350 (16-28% of total gaseous emissions), ethylene (8-23%), acetaldehyde (5-13%), acetylene (5-15%),
1351 propene (2-8%) and glyoxal (3-8%), with significant quantities of acrolein (<4%), benzene (<3%),
1352 1,3-butadiene (<3%), and toluene (<1%), while 16-42% of total non-methane volatile compounds
1353 remained unresolved. The sum of HCHO, ethylene, acetaldehyde, and propene may account for
1354 roughly 75% of the volatile organic compounds, while benzene, toluene, xylenes, and other
1355 substituted benzene compounds, oxygenates (acetone, glyoxal, and propanal), olefins (butene,
1356 pentene, hexane), and naphthalenes constitute the remaining 20% (Timko et al., 2010c). In addition
1357 to the numerous papers published, US Environmental Protection Agency (US EPA, 2009) also
1358 created a companion spreadsheet including data on speciated hydrocarbon from APEX projects.
1359 Figure 9 summarises the data from APEX campaigns in terms of profile (mass fraction) of the
1360 emitted hydrocarbons.

1361

1362 The total hydrocarbon EIs are highest at low power settings, where combustor temperatures and
1363 pressures are low and combustion is less efficient (Sutkus et al., 2001; Yelvington et al., 2007).
1364 UHC data provided by ICAO also confirm this behaviour for in-use TF engines (Figure 7).
1365 Similarly, many studies have reported the same behaviour for individual hydrocarbon species.
1366 Spicer et al. (1992; 1994) and Slemr et al. (2001) first reported that the emissions of many
1367 hydrocarbon species dropped at higher engine power by a factor of 20–50 and unburned fuel
1368 components disappeared. The EXCAVATE campaign (Anderson et al., 2006) also highlighted that
1369 most hydrocarbon species are strongly power dependent, with EIs at high thrusts dramatically lower
1370 than at idle. During APEX-1,2,3 campaigns, Knighton et al. (2007) observed that at engine power
1371 conditions significantly higher than 15% F_{00} , the engine combustion efficiency is close to 100%,

1372 resulting in hydrocarbon emissions often below the detection levels for many individual
1373 compounds. The inverse dependence of UHC upon thrust has a high relevance for air quality at
1374 airports, where idle and taxi phases are conducted at low thrusts and take up most of the time.
1375 Figure 8 shows that the cumulative UHC emission spans over two order of magnitude for in-use
1376 engines passing from idle to take-off during standardised LTO cycles.

1377

1378 Despite these interesting studies, the scientific literature still offers poor information on the
1379 hydrocarbon speciation and the few available data are often conflicting. For example, the potential
1380 changes in the hydrocarbon profiles at varying power are still unclear and deserve further
1381 investigation. Despite the large dependence of the magnitude of total UHC emitted from different
1382 engines, Knighton et al. (2009) observed that the ratios between the formaldehyde versus other
1383 hydrocarbon species were constant and independent of power settings. Although this result
1384 indicates constant hydrocarbon profiles with varying thrust, these results are inconsistent with other
1385 studies showing clear shifts of the hydrocarbon speciation with power. For example, during the
1386 EXCAVATE campaign, Anderson et al. (2006) observed that alkenes (mainly ethene) constituted
1387 more than 70% of the observed total NMHC emissions at idle, while at 61% F_{00} aromatic species
1388 (mostly toluene) accounted for over 50% of the total. There is currently a lack of information about
1389 the emitted hydrocarbons and this gap is mainly evident for emissions at power settings below the
1390 ICAO 7% idle. The behaviour and data for the most important classes of organics are discussed
1391 hereafter in separate sub-subsections.

1392

1393 **4.11.1 Methane**

1394 Methane (CH_4) is a radiatively active gas and is estimated to be 25 times more effective on a per-
1395 molecule level than CO_2 in terms of greenhouse effect at hundred-year time scales (Lelieveld et al,
1396 1998). Moreover, its roles in atmospheric chemistry to produce tropospheric ozone and
1397 stratospheric water vapour indirectly enhance its climate forcing effects. Although natural emissions

1398 from wetlands are largely recognised as dominant sources of methane at global scales,
1399 anthropogenic sources, such as energy, agriculture, waste and biomass burning can further
1400 contribute to its load in the atmosphere (Dlugokencky et al., 2011 and references therein). Most
1401 studies report that that turbine engines are not a significant source of CH₄ and have concluded that
1402 most engines tend to produce minor amounts of methane at idle and may consume it at higher
1403 engine power (Spicer et al., 1992, 1994; Vay et al., 1998; Slemr et al., 2001; Anderson et al., 2006;
1404 Santoni et al., 2011). Wiesen et al.(1994) examined methane emissions from different commercial
1405 jet engines (PW 305 and RB 211) under various flight conditions using different fuels and
1406 concluded that air traffic does not contribute significantly to the global budget of methane. Santoni
1407 et al. (2011) measured methane emissions from a CFM56-2C1 engine aboard a NASA DC-8
1408 aircraft and reported that the EI for CH₄ was (mean±standard deviation) 170±160 mg kg Fuel⁻¹ at
1409 4% and 7% F₀₀, while negative values (54±33 mg kg Fuel⁻¹) were reported for higher thrust
1410 settings, indicating consumption of methane by the engine.

1411

1412 **4.11.2 Alkanes, alkenes and alkynes**

1413 During the EXCAVATE campaign, Anderson et al. (2006) reported that the alkene species
1414 constituted over 90% of the observed total NMHC at idle but less than 20% at higher engine power
1415 settings. They also observed large decreases in alkane and alkene emissions with increasing engine
1416 power for various FSCs. In particular, EXCAVATE results showed that propylene underwent the
1417 most dramatic decrease, exhibiting a drop of mixing ratios by a factor ~280 from 7 to 61% F₀₀. In
1418 the same manner, isoprene dropped from ~2.5 ppbv to less than ~5 pptv (i.e., below the detection
1419 limit). On the other hand, these results reported decreases in alkane compounds which were much
1420 more modest, typically under a factor of 10. Schürmann et al. (2007) revealed that though isoprene
1421 was not directly found in emissions from kerosene refuelling, it was detected in considerable
1422 amounts in the aircraft exhaust which indicates that isoprene is most likely formed in the
1423 combustion process of a jet engine.

1424 **4.11.3 *Carbonyls***

1425 Due to their known adverse effects on human health, some carbonyls (formaldehyde, acetaldehyde,
1426 propionaldehyde and acrolein) have been included in the HAP list (Federal Aviation
1427 Administration, 2003). However, nowadays there is a gap in the current state of knowledge
1428 regarding the toxicity of many other aldehydes (including glyoxal, methylglyoxal and
1429 crotonaldehyde) which are detected in sizeable quantities in aircraft exhaust plumes and have
1430 potential toxic effects (Wood et al., 2008). APEX results (Kinsey, 2009) clearly showed that
1431 carbonyls generally account for most of the gaseous hydrocarbons emitted by common aircraft
1432 engines. Agrawal et al. (2008) reported that the major three contributors to carbonyl emissions are
1433 formaldehyde, acetaldehyde and acetone, and showed that carbonyl emissions are significantly
1434 higher during the idle mode than at higher thrusts. However, measurements of carbonyl EIs were
1435 also found to be very variable since they are sensitive to changes in ambient temperature
1436 (Yelvington et al., 2007; Knighton et al., 2007; Agrawal et al., 2008). Similar results were obtained
1437 for TS engines: Cain et al. (2013) observed that the EIs for the most prevalent aldehydes emitted at
1438 various engine power combinations were formaldehyde, acetaldehyde, and propionaldehyde and
1439 also reported a decrease with increasing engine power. The results of such engine tests seem to be
1440 confirmed by ambient measurements. For example, Fanning et al. (2007) and Zhu et al. (2011)
1441 reported that the time averaged concentrations of formaldehyde and acrolein were elevated at the
1442 Los Angeles International airport relative to a background reference site.

1443

1444 **4.11.4 *Aromatic compounds***

1445 Benzene, toluene, ethylbenzene, and *ortho*-, *meta*-, and *para*-xylenes are an important group of
1446 VOCs collectively known as BTEX. In urban environments BTEX are principally emitted by
1447 vehicle exhaust gases because of their presence in fuels, lubricating and heating oil, while minor
1448 sources include gasoline evaporation, use of solvents and paint, leakage from natural gas and
1449 liquefied petroleum gas. The adverse health effects of benzene are well known (e.g., WHO, 2000;

1450 Saillenfait et al., 2003; Pariselli et al., 2009, and reference therein) and it is included as a known
1451 human carcinogen by the IARC classification system. BTEX are highly reactive in the troposphere
1452 playing a key role in atmospheric chemistry as important photochemical precursors for tropospheric
1453 ozone and secondary organic aerosol generation (Atkinson, 2000; Atkinson and Arey, 2003).

1454

1455 Aromatic compounds are present in jet fuels, and can therefore be emitted as both unburned
1456 material and byproducts of incomplete hydrocarbon combustion, but also from fuel evaporation and
1457 refueling (Anderson et al., 2005; 2006). In this context, the benzene to toluene ratio (B/T) was often
1458 proposed to identify the fuel vs combustion origin of hydrocarbon mixtures. For example,
1459 Schürmann et al. (2007) observed that the B/T ratio at an airport is well below 1 for refuelling
1460 emissions and engine ignition while in the exhaust this value reaches up to 1.7. The US EPA (2009)
1461 mass fraction profiles (Figure 9) clearly show that BTEX account for ~4% of identified compounds,
1462 while other relevant aromatics (in order of decreasing mass fraction) are phenol, 1,2,4-
1463 trimethylbenzene, styrene, m-ethyltoluene and 1,2,3-trimethylbenzene. Generally, the literature
1464 shows large decreases in benzene and toluene emissions with increasing engine power, both for TF
1465 (Anderson et al., 2006) and TS engines (Cain et al., 2013). In particular, by studying the
1466 hydrocarbon emissions from a TS engine operating with conventional (JP-8), alternative and
1467 surrogate fuels, Cain et al. (2013) hypothesised that fuel composition and structure may play a
1468 significant role in the aromatic emissions of aircraft. They speculated that the propensity of the
1469 molecular structure of paraffins in fuels to produce benzene or toluene was observed to follow
1470 cycloparaffin > iso-paraffin > *n*-paraffin. This study also attempted to depict the chemical processes
1471 at the basis of their observations and hypothesised that iso- and *n*-paraffins must first undergo either
1472 ring closure or decomposition to combustion/pyrolytic intermediates prone to ring formation (e.g.,
1473 propargyl radicals and propylene) to ultimately form cyclic and aromatic compounds. In addition,
1474 Cain et al. (2013) reported that an increased branching ratio of iso-paraffins resulted in higher

1475 production rates of the C₃-intermediates, which further contribute to ring/aromatic formation and
1476 growth.

1477

1478 **4.11.5 Polycyclic aromatic hydrocarbons**

1479 Among the large number of hydrocarbon species emitted by aircraft engines, the polycyclic
1480 aromatic hydrocarbons (PAHs) deserve particular attention because most congeners are known,
1481 probable or possible human carcinogens (WHO, 2000; Armstrong et al., 2004; IARC, 2010) and
1482 because of their ubiquitous presence in the urban atmosphere (Ravindra et al., 2008; Zhang and
1483 Tao, 2009). PAH are semi-volatile and partition between the gaseous and particulate phases; lighter
1484 PAHs (2 to 3 aromatic rings) are present almost exclusively in the vapour-phase, whereas PAHs
1485 with higher molecular weights (>4 rings) are almost totally adsorbed on particles. Although PAHs
1486 may undergo oxidation by several atmospheric oxidants, their potential for long range transport
1487 cannot be disregarded (e.g., Keyte et al., 2013).

1488

1489 Agrawal et al. (2008) showed that lighter congeners such naphthalene and its 1-methyl and 2-
1490 methyl derivatives contribute strongly to the total PAH mass in various aircraft (TF) emissions at
1491 differing thrust modes. Moreover, they also reported that the EI(naphthalene) increased as power
1492 increased from idle mode falling off as the engine operated at the highest power. Chen et al. (2006)
1493 characterised the PAH emissions of the TS engine of a helicopter at five power settings and
1494 reported a mean total PAH concentration in the exhaust of 843 $\mu\text{g m}^{-3}$, with a maximum of 1653 μg
1495 m^{-3} emitted during ground idle. The emission level of total PAHs during a complete LTO cycle was
1496 estimated to be 1.15 g PAHs LTO⁻¹. Even if the results provide evidence for high mass
1497 concentrations of total emitted PAH, the speciation revealed that lighter congeners, which have
1498 generally lower carcinogenic potencies, were dominant: 59.7% of total PAHs emissions were made
1499 up of naphthalene, 37.8% of three-ring congeners, while the remaining 2.5% of PAHs had four- to

seven-rings. The emission factor revealed U-shaped behaviour: maximum at idle (50%), minimum at fly idle (67%) and increasing until max thrust (100% F_{00}).

1502

Although the PAH pollution at airports can be overwhelmed by external sources, such as vehicular traffic and industrial emissions, a number of studies have indicated airport emissions cannot be neglected. Cavallo et al. (2006) measured the concentrations of 23 PAH in three areas (airport apron, building and terminal/office) of a major Italian airport (Fiumicino, Rome). The airport apron was found to be suffering the highest levels of total PAHs ($27.7 \mu\text{g m}^{-3}$) with a prevalence of 2–3 ring PAH such as methylnaphthalenes and acenaphthene presumably associated with jet fuel combustion. However, they also showed that PAH levels were lower than the threshold limit value proposed for occupational exposure by ACGIH (0.2 mg m^{-3}). Similar results were obtained by Zhu et al. (2011), who observed that the semi-volatile PAHs (from phenanthrene to chrysene) were consistently higher at both blast fence and downwind sites from the take-off runway of Los Angeles airport than at a background site. This study also indicated naphthalene as the most abundant gas-phase PAH (80–85% of the total PAHs).

1515

1516 **4.11.6 Organic sulfur, nitrogen and chlorinated species**

Since jet fuels contain variable FSC, some organic sulfur species may form during combustion. Anderson et al. (2006) measured the emissions of OCS, CS_2 and dimethyl sulphide (DMS) from a RB211-series TF engine at varying engine power and burning two different FSC fuels. Results showed no consistent trends for OCS and CS_2 with varying thrust settings and suggested that the sources of those gases are insensitive to the FSC. In contrast, this study revealed that levels of DMS are dramatically reduced from approximately ambient levels at idle to near the instrument detection limit as engine power is increased and speculated that ambient DMS is essentially burned (oxidised) out of the exhaust stream at combustor temperatures associated with high engine power.

1525

1526 The presence of organic nitrogen species in aircraft exhaust may derive from the presence of
1527 nitrogen in fuels and from the potential reaction between alkanes and NO_x within the exhaust
1528 plume. During the EXCAVATE campaign, alkyl nitrate species were observed in exhaust plumes
1529 with methyl nitrate, iso-propyl nitrate, and 2-butyl nitrate accounting for 80–90% of the total N-
1530 containing organic species (Anderson et al., 2006). In particular, methyl nitrate was observed to
1531 follow U-shaped curves of EI vs. fuel flow, with minimum emissions at mid-range thrust, slightly
1532 increased emissions at low thrust and strongly increased at higher powers.

1533

1534 Chlorinated organic compounds can form in aircraft exhaust as by-products of fossil fuel
1535 combustion in the presence of chlorine. Chlorine can be present in fuels because refineries can use
1536 salt driers to remove water from fuels (Anderson et al., 2006), and in certain circumstances may be
1537 present in ambient air as sea salt, such as in coastal environments. Despite the lack of available data
1538 in the literature, there is no evidence to date that chlorinated compounds are produced by aircraft
1539 engines. For example, Agrawal et al. (2008) observed that the emissions of dioxins from various
1540 aircraft engines are below the detection limit.

1541

1542 **4.12 Chemi-ions**

1543 Aircraft exhausts also contain gaseous ions, the so called chemi-ions (CIs), have been measured in
1544 several studies (e.g., Reiner and Arnold, 1993;1994; Arnold et al., 1998b; Yu and Turco, 1997;
1545 Kiendler and Arnold, 2002; Eichkorn et al., 2002; Haverkamp et al., 2004; Sorokin et al., 2004;
1546 Miller et al., 2005; Anderson et al., 2005). Their formation was also found in various mobile
1547 sources (e.g., Seigneur, 2009) and is attributed to the radical–radical reactions during combustion
1548 processes. Once emitted, CIs may evolve chemically via ion-ion recombination and ion-molecule
1549 reactions involving trace gas molecules present in the exhaust (Kiendler and Arnold, 2002) and may
1550 act as aerosol precursors (Sorokin and Mirabel, 2001; Eichkorn et al., 2002). Starik (2008) provides
1551 a scheme of ion formation in hydrocarbon flames and inside the combustor.

1552 Relatively high number concentrations of CIs have been measured: in the SULFUR experiments
 1553 (Schumann et al., 2002 and reference therein) 10^9 ions cm^{-3} were reported at ground level, i.e., of
 1554 the order of 10^{17} CIs kg Fuel^{-1} , but it was also reported that CIs decrease rapidly with increasing
 1555 plume age (Arnold et al., 2000; Sorokin and Mirabel, 2001). Haverkamp et al. (2004) measured EI
 1556 for the total (positive and negative) ions of 1.2×10^{16} - 2×10^{16} CIs kg Fuel^{-1} and observed number
 1557 concentrations of the same order of magnitude for both negative and positive ions: negative CIs
 1558 varied from 6×10^7 and 2.1×10^8 molecules cm^{-3} , while positive ions ranged from 4×10^7 to 1.7×10^8
 1559 molecules cm^{-3} . About 50% of the measured ions have masses heavier than 100 amu and the most
 1560 massive ions show masses up to 1500-3000 amu, depending on the fuel flow (thrust) and FSC
 1561 (Haverkamp et al., 2004). Schumann et al. (2002) reported masses also exceeding 8500 amu.
 1562 Identified negative CIs include many organic ions and cluster ions containing sulfuric acid, e.g.,
 1563 $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_n$, $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_n(\text{SO}_3)_m$ ($n < 3$, $m = 0, 1$), $\text{NO}_3^-(\text{HNO}_3)_m$ and $\text{HSO}_4^-(\text{HNO}_3)_m$
 1564 ($m = 1, 2$). Kiendler and Arnold (2002) further reported a low stability of $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_n$ ($n \geq 3$)
 1565 against thermal detachment of H_2SO_4 at high temperatures, indicating the presence of gaseous
 1566 H_2SO_4 in exhaust plumes. Positive CIs are mostly oxygen-containing organic compounds
 1567 (Schumann et al., 2002) and considering the heavy masses of most CI, Haverkamp et al. (2004) also
 1568 hypothesized the presence of large organic molecules, such as PAHs.
 1569

1570 The generation of CIs in the combustor, their physico-chemical characteristics and the changes
 1571 occurring along with plume aging are not yet well understood and merit further investigation as
 1572 these ions may play a key role in the formation of numerous volatile aerosol particles (e.g., Yu and
 1573 Turco, 1997; Arnold et al., 2000; Sorokin and Mirabel, 2001; Haverkamp et al., 2004; Miller et al.,
 1574 2005).
 1575
 1576
 1577

1578 **4.13 Particulate Matter**

1579 Particulate matter (PM) is emitted by a great variety of both natural and anthropogenic sources. The
1580 latter include a large variety of anthropogenic processes, which emit particles with very different
1581 chemical composition and physical properties. Nowadays, PM composition and sources have been
1582 extensively investigated in a large number of different environments (e.g., Viana et al., 2008;
1583 Harrison et al., 2012; Amato et al., 2013). However, few data on PM emissions are historically
1584 available for aircraft engines (Wayson et al., 2009, Kinsey et al., 2011). In addition, ICAO has not
1585 yet defined any emission standard for PM to be applied during LTO cycles and is therefore
1586 interested in setting a certification limit for this pollutant to address related air quality and climate
1587 issues (Kinsey, 2009). In this context, there are some current programmes aiming to describe the
1588 PM emissions from aircraft engines, e.g., the Society of Automotive Engineers (SAE) E-31
1589 Committee is developing a standard PM test method for aircraft engine certification (SAE, 2009).

1590

1591 Despite a number of studies which have been published recently on PM emissions from gas turbine
1592 engines from both a physical and a chemical point of view (e.g., Corporan et al., 2008; Whitefield et
1593 al., 2008; Herndon et al., 2008; Agrawal et al., 2008; Westerdahl et al., 2008; Kinsey et al., 2010;
1594 2011), current data on aircraft-generated PM are still wholly inadequate and many open questions
1595 wait to be addressed. This gap appears to be a pressing issue because many epidemiological studies
1596 have found a strong correlation between the exposure to PM and some significant adverse human
1597 health effects (e.g., Pope and Dockery, 2006; Valavanidis et al., 2008; Polichetti et al., 2009;
1598 Karakatsani et al., 2012; Anderson et al., 2012; Heal et al., 2012; Martinelli et al., 2013). PM
1599 inhalation can affect morbidity and can lead to an increase in hospital admissions, and is
1600 significantly associated with mortality and to a substantial reduction in life expectancy (Pope et al.,
1601 2009; Hoek et al., 2010; Sapkota et al., 2012; Raaschou-Nielsen et al., 2013).

1602

1603

1604 **4.13.1 Volatile and non-volatile PM**

1605 PM generated from aircraft engines can be classified into two major fractions: non-volatile and
1606 volatile PM (e.g., Kinsey et al., 2009; Presto et al., 2011), while the combination of both volatile
1607 and non-volatile PM is commonly referred as total PM. Non-volatile PM is directly emitted by
1608 engines and is mainly composed of graphitic/elemental/black carbon with traces of metals, which
1609 are stable at the high temperatures and pressures normally reached in the exhaust plumes. Volatile
1610 PM is instead formed through the gas-to-particle partitioning and conversion processes of sulfur and
1611 various organic gases (Robinson et al., 2010; Timko et al., 2010b), which occur after the emission
1612 in the near-field plume downstream of the engine (Kinsey et al., 2011). Since the most volatile PM
1613 components are partitioned into the gas- and particulate-phases, their behaviour is sensitive on the
1614 changes in the environmental conditions with respect to the near-plume and in any case many
1615 compounds can remain in equilibrium between the two phases. This component is therefore very
1616 sensitive to the sampling conditions (Wey et al., 2006; Wong et al., 2011; Presto et al., 2011). In
1617 particular, the organic component of the volatile PM undergoing partitioning between the two
1618 phases is named organic aerosol (OA) and can be composed of a large number of different
1619 hydrocarbon classes. Moreover, as the reactive compounds can be affected by oxidation by a
1620 number of atmospheric oxidant species (mainly hydroxyl, nitrate radicals and ozone), it can be
1621 expected that the composition and the quantity of volatile PM changes progressively away from the
1622 plume, after natural cooling, dilution and chemical processes occur in the atmosphere. Many
1623 hydrocarbons of high volatility, such as BTEX, low molecular weight PAHs, alkanes and many
1624 others, may be easily oxidised to species with substantially lower volatilities (Kroll and Seinfeld,
1625 2008) and, thus, may act as precursors for the formation of the secondary organic aerosol (SOA).
1626 The formation and the properties of the SOA, including their gas/particle partitioning, are an intense
1627 area of research (e.g., Pandis et al., 1992; Pankov, 1994; Odum et al., 1996; Kroll and Seinfeld,
1628 2008; Hallquist et al., 2009) and the common way to describe the partitioning of a constituent *i*

1629 between the gas- and the condensed- phases with mass concentration C_{OA} can be described by a
1630 partitioning coefficient, ξ_i :

1631
$$\xi_i = 1/[1+(C_i^*/C_{OA})]$$

1632 where C_i^* is the effective saturation concentration of the compound, i.e. a semi-empirical property
1633 describing the partitioning of complex mixtures. Donahue et al. (2009) proposed three different
1634 classes of compounds on the basis of their C^* values: (i) the low volatility organic compounds,
1635 showing C^* from 10^{-2} to $10^{-1} \mu\text{g m}^{-3}$ and mostly remaining in the condensed phase under common
1636 atmospheric conditions; (ii) the SVOCs, exhibiting C^* between 10^0 and $10^2 \mu\text{g m}^{-3}$ and undergoing
1637 significant partitioning and (iii) the intermediate volatility organic compounds (IVOCs), having C^*
1638 in the order of magnitude of 10^3 — $10^6 \mu\text{g m}^{-3}$, which are almost entirely in the gas-phase. Recently,
1639 some studies have pointed out that most hydrocarbons emitted by aircraft engines are thought to be
1640 important SOA precursors (Miracolo et al., 2011; Presto et al., 2011), being in the IVOC and SVOC
1641 classes. However, the potential of hydrocarbons emitted by aircraft exhaust to form secondary
1642 components is currently poorly understood.

1643

1644 **4.13.2 Particulate mass**

1645 Generally, the emission indices of PM mass range from approximately 10 to 550 mg PM kg Fuel⁻¹
1646 (Kinsey, 2009). U-shaped curves of PM emissions versus thrust are commonly reported in the
1647 literature, showing elevated emissions at low power settings, a decrease to a minimum at midrange
1648 power, and then an increase at high or full power (Whitefield et al., 2008; Kinsey, 2009; Kinsey et
1649 al., 2010; 2011). Agrawal et al. (2008) noted a 10 to 40-fold increase in the EI(PM) as the engine
1650 power increased from idle to climb thrust. However, there are deviations from this behaviour: the
1651 PM mass emission indices at varying thrusts have been shown to depend on various factors,
1652 including engine families, technology, FSC, operating power, cold and warm engine conditions and
1653 environmental conditions (e.g., Kinsey, 2009) and real-time emission rates for PM for a typical TF
1654 engine have revealed significant PM spikes during changes in power settings (Agrawal et al., 2008).

1655 The measurements of PM from aircraft exhaust are heavily dependent on the adopted methodology
 1656 (e.g., Presto et al., 2011). Since the volatile PM may undergo rapid changes in time and space, the
 1657 sampling protocol, such as the distance from the engine exit, and other parameters having
 1658 implications on the aging of plumes play a key role in the mass of sampled particles. In addition, the
 1659 environmental conditions (e.g., temperature, humidity, sunlight, wind, etc.) can also affect PM
 1660 mass, particularly through the potential for particle formation, coagulation, and growth (e.g.,
 1661 Herndon et al., 2005). Timko et al. (2010b) reported that soot is the only type of particle detected at
 1662 the engine exit plane, while volatile particles are only detected downwind (15–50 m) due to the
 1663 nucleation of sulphate and organic materials in the cooling exhaust plume. Kinsey et al. (2010)
 1664 indicated that a variable amount (40% to 80%) of the total PM can be composed of volatile matter,
 1665 mainly in the form of sulfur and organics. Lobo et al. (2012) measured the specific PM emissions
 1666 during normal LTO operations at a distance of 100-300 m downwind of an active taxi-/runway at
 1667 the Oakland International Airport and reported EI(PM) between 100 and 700 mg PM kg Fuel⁻¹
 1668 under both the idle/taxi and take-off conditions for various aircraft/engine combinations.

1669

1670 **4.13.3 Particle number concentration**

1671 During the APEX campaigns, the observed EI(#) varied from approximately $1 \cdot 10^{15}$ to $1 \cdot 10^{17}$
 1672 particles kg Fuel⁻¹ (Kinsey, 2009; Kinsey et al., 2010) and are therefore comparable on a per unit
 1673 fuel burn basis to the number of particles generated from other combustion sources, such as ship
 1674 emissions, biomass burning and forest fires (Kumar et al., 2013). Generally most TF engines tested
 1675 during APEX projects exhibited EI(#) strongly correlated with fuel flow (Kinsey et al., 2010), with
 1676 higher EI at low power settings following a logarithmic relationship of EI(#) to thrust:

$$1677 \quad EI(\#) = m \cdot [\ln(\text{fuel flow})] + b$$

1678 where m represents the slope of the regression line with values ranging from $-2 \cdot 10^{15}$ to $-3 \cdot 10^{16}$ and b
 1679 is the intercept of the regression line varying from $2 \cdot 10^{16}$ to $2 \cdot 10^{17}$ (Kinsey, 2009). Similarly to
 1680 EI(PM) the particle number indices were however observed to be sensitive to engine technology,

1681 FSC, operating power and environmental conditions: Kinsey (2009) also reported a completely
 1682 different behaviour for a TJ engine (CJ610-8ATJ), with EI(#) lower at idle and relatively constant at
 1683 higher F_{00} .
 1684

1685 It was shown that EI(#) tends to increase moving away from the engine exit plane. EXCAVATE
 1686 results (Anderson et al., 2005) reported increases by a factor of 10 at 25 to 35 m than at 1 m
 1687 downstream of the exhaust plane. Timko et al. (2010b) further observed differences in particle
 1688 number emissions sampled at engine exit plane and downwind (15-50 m) of the engine. They
 1689 reported that soot is the main species detected at the engine exit plane, while the nucleation of
 1690 volatile particles in the cooling exhaust gases measured downwind further led to increases in the
 1691 particle number of 1-2 orders of magnitude.
 1692

1693 Cheng and Corporan (2010) reported particle number emissions from military engines operated
 1694 with JP-8 fuel in various thrust settings. They observed that a common TF engine emits increasing
 1695 number of particles at increasing thrust with particle number emission indices of $5.5 \cdot 10^{15}$, $5.3 \cdot 10^{15}$,
 1696 $9.6 \cdot 10^{15}$, and $8.9 \cdot 10^{15}$ particles kg Fuel^{-1} for the idle, 80%, 90% and 95% power setting,
 1697 respectively. A inverse pattern with decreasing emissions at increased power settings was instead
 1698 reported for a common TP engine equipping the widespread used military cargo C-130 Hercules:
 1699 averaged EI were $1.8 \cdot 10^{16}$, $1.4 \cdot 10^{16}$, $1.4 \cdot 10^{16}$, $1.0 \cdot 10^{16}$, and $1.2 \cdot 10^{16}$ particles kg-fuel^{-1} for 4%, 7%,
 1700 20%, 41% and max thrusts, respectively. This study also examined two common TS engines used in
 1701 most helicopters and aircraft and reported increasing emissions of particles with increasing thrust:
 1702 $3.1 \cdot 10^{15}$ (idle), $3.3 \cdot 10^{15}$ (75%) and $5.5 \cdot 10^{15}$ (max thrust) particles kg-fuel^{-1} and $1.1 \cdot 10^{14}$ (idle)
 1703 $1.8 \cdot 10^{15}$ (75%) and $3.0 \cdot 10^{15}$ (max thrust), respectively. Similar results were observed by Cain et al.
 1704 (2013) in a TS engine burning various types of fuel: JP-8 fuel emissions were between 10^{15} and 10^{16}
 1705 particles kg-fuel^{-1} , while emissions from other alternative and surrogate fuels were 1 to 2 order of
 1706 magnitude lower.

Measurements of EI(#) at airports indicated similar results. Lobo et al. (2012) measured the specific PM emissions during normal LTO operations at a distance 100-300 m downwind of an active taxi-/runway at the Oakland International Airport and associated the data with various aircraft/engine combinations. They observed similar EI(#) for both idle/taxi ($7 \cdot 10^{15}$ - $3 \cdot 10^{17}$ particles kg Fuel⁻¹) and take-off ($4 \cdot 10^{15}$ - $2 \cdot 10^{17}$ particles kg Fuel⁻¹) phases. Klapmeyer and Marr (2012) reported that the EI(#) for in-use aircraft at a regional airport varied from $1.4 \cdot 10^{16}$ to $7.1 \cdot 10^{16}$ particles kg Fuel⁻¹ and observed slightly higher concentrations during taxi phases than during take-offs.

1714

The beneficial effects of alternative fuels upon particle emissions are nowadays under discussion. Although this review does not focus on such effects, it is interesting to note that some studies have highlighted potential positive effects on the EI(#) and EI(PM). For example, Lobo et al. (2011) reported reduced emissions of PM number emissions of about one third using 50% FT/50% Jet-A1 blend instead of Jet-A1.

1720

1721 **4.13.4 Size distributions**

Size distributions of airborne particles influence their residence time and dispersion (Allen et al., 2001). In addition, the dimensions of particles are directly related to their emission sources, as mechanically generated particles (e.g., wind-blown dust, sea spray) are generally largest than 1 µm, while combustion-generated (high-temperature processes, traffic, many industrial activities) are typically smaller than 1 µm (e.g., Lewis and Schwartz, 2004; Seinfeld and Pandis, 2006; Ning and Sioutas, 2010). Ultrafine particles (UFPs, diameter <100 nm) typically constitute ~90% or more of particle number count in areas influenced by vehicle emissions (Morawska et al., 2008). UFPs have larger surface area per unit mass with respect to larger particles and can potentially contain high proportions of organic material such as polycyclic aromatic hydrocarbons. Moreover, UFPs can penetrate deeper into the respiratory tract and into cells possibly posing an elevated risk for human

1732 health (Oberdorster et al., 2004; Delfino et al., 2005; Bräuner et al., 2007; Belleudi et al., 2010;
1733 Knibbs et al., 2011).

1734

1735 A large number of studies (e.g., Herndon et al., 2005; Wey et al., 2007; Westerdahl et al., 2008;
1736 Cheng et al., 2008; Mazaheri et al., 2009; Dodson et al., 2009; Kinsey, 2009; Kinsey et al., 2011;
1737 Zhu et al., 2011; Presto et al., 2011; Hsu et al., 2013) have provided evidence that AEs may lead to
1738 increased concentrations of UFPs. However, the nature of semi-volatile compounds emitted by
1739 aircraft, the possible mechanisms of secondary aerosol formation and the dilution effect, make it
1740 difficult to associate a measured size distribution with a specific source. Studies performed at the
1741 exhaust exit-plane or directly downstream of the engine cannot usefully be compared with data
1742 obtained in ambient air sampled at airports. However, even if differences and limitations exist,
1743 some trends and recurring modes have been identified in most studies.

1744

1745 A study by Schumway (2002) used scanning electron microscopy to analyse individual particles
1746 emitted from military engines and reported predominant particles with dimensions ranging from 22
1747 to 120 nm. It was observed that emitted particles were discrete at low thrust (approach and idle),
1748 while they tended to agglomerate at higher power (intermediate and military modes). Similar results
1749 have recently been reported by Mazaheri et al. (2013), who analyzed the aircraft emissions during
1750 normal takeoff and landing operations at an international airport by using the transmission electron
1751 microscopy technique. They reported particles in the range of 5–100 nm in diameter with a
1752 dominant nucleation mode (18–20 nm) and semisolid spherical shapes. Nowadays most studies
1753 measure particle size distributions using automatic instruments, such as scanning mobility particle
1754 sizers (SMPS), electrical low pressure impactors (ELPI), and differential mobility spectrometers
1755 (DMS). A comprehensive review of these devices is provided elsewhere (Kumar et al., 2010).
1756 Anderson et al. (2005) reported that exhaust exit-plane measurements on engines mounted in test
1757 cells and B757 aircraft in run-up facilities produce of the order of 10^{15} soot particles per kg of fuel

1758 burned with a mean mass diameter of 40 to 60 nm. Using an improved version of the nanometre
1759 aerosol size analyser (nASA), they also reported that the aerosol size distribution at 1 m from a
1760 B757 engine is a combination of volatile and non-volatile particles with a bimodal distribution. The
1761 first (non-volatile) mode was measured by heating the aerosol to 300°C before analysis with the
1762 nASA and was found to be around 20 nm; this mode was thought to be primarily composed of soot
1763 and other components including zinc, aluminium, and titanium which are from the abrasion of
1764 engine components or the trace metal impurities in the fuel. The second (volatile) mode was
1765 observed at 7 nm and comprised particles that vaporise below 300°C.

1766

1767 During the APEX campaigns (e.g., Wey et al., 2007; Kinsey, 2009; Kinsey et al., 2010), the particle
1768 size distributions of the emissions were generally found to be unimodal and log-normally
1769 distributed, with electrical mobility diameters ranging from ~3 nm to >100 nm and a geometric
1770 number mean diameter (GMD) of ~10–35 nm. A slight dependence of GMD on thrust was
1771 detected, with GMD of 10–20 nm at low fuel flow rates, a decrease at mid-power and then an
1772 increase at higher thrust. These studies also reported the presence of a prominent nucleation mode
1773 mainly on samples collected farther from the engine exit (30 m) with respect to gases sampled at 1
1774 or 10 m. This second mode was attributed to the secondary aerosol generation caused by the
1775 expansion and cooling of the exhaust plume and is composed of sulfuric acid and low-volatility
1776 hydrocarbons (Wey et al., 2007). APEX results detected changes in both the GMD and related
1777 geometric standard deviation (GSD) of the particle size distributions at varying engine and fuel
1778 type, thrust, and environmental conditions.

1779

1780 While APEX reported size distributions for commercial in-use airliner engines, we report data from
1781 other studies on differing engine types and technologies. Rogers et al. (2005) showed that the
1782 particles measured in the exhaust of two military engines (a FT with afterburner and a TS) were
1783 unimodally distributed with peaks at 20–40 nm. Cheng et al. (2008) observed that the particle

number size distributions downstream of a C-130 Hercules showed peaks between 50 and 80 nm for engine power settings ranging from idle to maximum thrust. They also observed a clear trend of increasing particle diameter with increasing engine power setting and distance from the engine exit. Cheng et al. (2008) detected the presence of another peak corresponding to the lower instrumental limit, presumed to be an additional mode below 20 nm. Cheng and Corporan (2010) reported unimodal size distributions for military turbofan, turboprop and turboshaft emissions sampled at the engine exhaust plane. They observed that both the total particle number concentration and GMD increased as the engine power increased for all tested engines. In particular, the observed GMD ranged from 55 nm (at idle) to 85 nm (at 95% F_{00}) in turbofan, from 51 nm (at idle) to 67 nm (at max thrust) in turboprop and from 20 nm (at idle) to 42 nm (at max thrust) in a turboshaft engine.

1794

1795 **4.13.5 *Changes of particle number and size after the dilution of plumes***

1796 The effects of the aircraft-related emissions of UFP at airports have received increasing attention in
1797 recent years and some studies have demonstrated a clear dependence of UFP concentrations and
1798 size distributions upon aircraft operations. In addition, UFP measurements upwind and downwind
1799 of airports are of particular importance because they are performed under ambient conditions, i.e.
1800 after the plume has been diluted by air and the particle coagulation and gas-to-particle condensation
1801 processes have occurred.

1802

1803 Hu et al. (2009) studied the effect of aircraft movements in a neighbourhood adjacent to the
1804 regional airport of Santa Monica and observed that spikes in the particle number concentration
1805 related to the take-off phase were 440 times elevated above background and reached 2.2×10^6
1806 particles cm^{-3} . At a site located at the blast fence of Los Angeles International Airport, Zhu et al.
1807 (2011) reported that total UFPs counts exceeded 10^7 particles cm^{-3} during take-offs. This study
1808 further investigated temporal profiles in particle concentration of 30 nm mobility diameter
1809 (corresponding to the mean geometric mode of emitted particles) due to isolated aircraft take-off

1810 events: dramatic increases of particle concentrations (from $1.6 \cdot 10^3$ to $1.7 \cdot 10^4$ particles cm^{-3}) were
 1811 reported when aircraft engines are accelerated to the 100% thrust power for take-off, followed by
 1812 decreases of number concentrations showing an exponential decay. Similar findings have been
 1813 reported by Hsu et al. (2012), who observed that departures of jet engine aircraft on a runway may
 1814 contribute to $1 \cdot 10^3$ to $7 \cdot 10^4$ particles cm^{-3} . The same authors further revealed significant higher
 1815 increases of UFP at Los Angeles International airport (Hsu et al., 2013) due to the LTO activity:
 1816 $2 \cdot 10^6$ – $7 \cdot 10^6$ particles cm^{-3} increase at a monitor at the end of the departure runway,
 1817 $8 \cdot 10^4$ – $1.4 \cdot 10^5$ particles cm^{-3} at a site 250 m downwind from the runway.
 1818
 1819 Changes in the particle size distributions can also occur after plumes are diluted in ambient air due
 1820 to coagulation. However, most studies have shown that particle size distributions at airports are
 1821 comparable with those measured during engine tests. Air monitoring carried out in the surroundings
 1822 of the Los Angeles International Airport found that the upwind site was dominated by particles of
 1823 approximately 90 nm diameter whereas downwind sites were dominated by finer particles, peaking
 1824 at approximately 10–15 nm (Westerdahl et al., 2008), which corresponds to the size reported during
 1825 APEX campaigns for many in-use engines (Kinsey et al., 2010). Similarly, Fanning et al. (2007)
 1826 and Zhu et al. (2011) reported very high number concentrations of UFPs collected at the blast fence
 1827 site, with the highest numbers found at a particle size of approximately 14 nm. The same study
 1828 further observed that the UFP number concentrations measured in a residential community
 1829 approximately 2–3 km downwind of the airport were intermediate in concentration between the
 1830 airport runway and the background reference site. This finding was associated with aircraft take-off
 1831 activities and the authors noted the significant exposure and possible health implications for people
 1832 living near the airport. Mazaheri et al. (2009) revealed that size distributions exhibit similar
 1833 modality during all phases of the LTO cycles with particles predominantly in the range of 4–100 nm
 1834 in diameter. This latter study also reported two distinct modes: a nucleation mode at diameters <30
 1835 nm observed in all LTO modes and an accumulation mode between 40 and 100 nm more

1836 pronounced during take-offs. While the nucleation mode exhibited the highest number
1837 concentration of all modes, the accumulation mode dominated the particle mass size distributions.
1838 Lobo et al. (2012) measured the specific PM emissions during normal LTO operations at a distance
1839 of 100-300 m downwind of an active taxi-/runway at the Oakland International Airport and
1840 associated the data with various aircraft/engine combinations. The size distributions were typically
1841 bimodal with a nucleation mode composed of freshly nucleated PM and an accumulation mode
1842 mostly made up of soot with some condensed volatile material. These observations closely parallel
1843 the mechanisms and size distribution of particles in diesel exhaust (Harrison et al., 2011).

1844

1845 **4.14 Chemical Composition of PM**

1846 Although the chemical composition of PM may include most of the periodic table of the elements
1847 and many thousands of different organic compounds, it is principally composed of few major
1848 components, which usually represent several percent of the total mass of particles, and some of
1849 those may remain in thermodynamic equilibrium between gaseous and particle phases. The
1850 particulate matter emitted directly by aircraft is mostly composed of soot (e.g., Anderson et al.,
1851 2005; Timko et al., 2010b), while sulphate and semi-volatile hydrocarbons may further coat the
1852 particles after the plume dilution. However, aircraft PM may also contain traces of metals and ions,
1853 which are mainly the result of: (i) fuel impurities; (ii) corrosion and wear of mechanical
1854 components of engines; (iii) pre-existing PM drawn in the combustor. The following sub-
1855 subsections discuss the various components separately.

1856

1857 **4.14.1 Carbonaceous PM**

1858 Carbonaceous PM consists of a complex mixture of elemental carbon (EC) and organic carbon
1859 (OC) (jointly referred to as soot) and commonly accounts for a large fraction of ambient fine
1860 particle mass in both rural and urban environments. Soot is primarily generated by incomplete
1861 combustion processes through the pyrolysis of organic fuels used in combustion processes. Many

1862 studies have discussed the various types of such particles; however there are still controversies and
1863 open discussion about the terminology to adopt. The terms used to identify the various fractions of
1864 carbonaceous aerosols, such as soot, black carbon (BC), elemental carbon (EC), equivalent black
1865 carbon and refractory black carbon are mainly associated with the corresponding measurement
1866 methods (e.g., Pöschl, 2003; Andreae and Gelencsér, 2006; Bond and Bergström, 2006; Kondo et
1867 al., 2011; Buseck et al., 2012; Long et al., 2013; Novakov and Rosen, 2013) and more generally
1868 refer to the most refractory and light-absorbing component of carbonaceous combustion particles,
1869 even if the underlying definitions and measurement methods are different (Petzold et al., 2013).
1870 Without going into the merits of this discussion, this section provides an overview of the data
1871 concerning the carbonaceous fraction and the terms used (soot, BC and EC) are the same as
1872 reported by the original authors. In any case, Lee et al. (2010) indicated that BC is often used
1873 interchangeably with soot in the literature relating to aircraft emissions, although in the strictest
1874 sense they are different.

1875

1876 The airliners of 1960s and 1970s emitted visible and dark exhaust plumes, especially during take-
1877 off. In recent decades, a great effort has been made by most engine manufacturers to reduce such
1878 emissions, which consisted mainly of soot and organics, and nowadays most modern airliners do
1879 not emit visible plumes. However, soot is still the primary form of non-volatile PM emitted by jet
1880 engines (e.g., Timko et al., 2010b), even if its contribution represents only few percent of the global
1881 atmospheric BC emission (Hendricks et al., 2004).

1882

1883 From a morphological point of view, soot particles emitted by aircraft engines have nearly spherical
1884 shapes with lognormal size distributions peaking at 30–60 nm (Petzold et al., 2003, 2005a;
1885 Popovicheva et al., 2004). However, once emitted soot particles quickly build complex
1886 agglomerates causing a second mode of larger particles between 100 and 500 nm, which are totally
1887 amorphous (Petzold et al., 1998; Popovicheva et al., 2000; 2004; Demirdjian et al., 2007). Despite

the structural characteristics of soot being of primary importance in relation to its atmospheric properties, there is a lack of experimental data on microstructure, composition and hygroscopicity of original soot emitted from aircraft engines. Some studies conducted at cruise height (Kärcher et al., 1996; Gleitsmann and Zellner, 1998) have assumed that all the soot particles in exhausts are hydrophobic. Demirdjian et al. (2007) used a combination of several analytical methods to study the microstructure and the composition of soot agglomerates sampled in an aircraft engine combustor and reported that soot was in two main fractions having quite different physicochemical properties. A major fraction of particles was found to be made up of amorphous carbon with small amounts of oxygen, sulfur and iron and was rather hydrophobic, while a second fraction was characterised by various structures and a large amount of impurities and was highly hydrophilic. Vander Wal et al. (2010) compared the physical structure and the chemical composition of soot produced by different sources, including a modern TF engine, using high resolution transmission electron microscopy and X-ray photoelectron spectroscopy. The results showed that some physical characteristics of jet engine soot, such as the lamella length distributions, are intermediate between soot produced by other sources such as wildfires and diesel, while other characteristics are singular. Jet soot was reported to have the highest sp^3 carbon content, in fact higher than the sp^2 (graphitic) content, the greatest oxygen content in the form of phenolic and carbonyl groups and the widest range of hetero-elements, including S, Na, N, Zn, Ba.

1906

From a chemical point of view, soot is mainly made up of graphitic BC (Petzold et al., 1999; Popovicheva et al., 2004), but some particles can be also coated with organic materials and sulfur species (e.g., Petzold et al., 2003). For example, the hygroscopic properties of jet engine combustion particles have been investigated in several rig-tests and results have confirmed that the water uptake by combustion particles is generally independent of combustor operating conditions, but increases significantly with increasing FSC level, which is attributed to an increasing amount of sulfuric acid adsorbed on the particles (Gysel et al., 2003). The uptake of sulfuric acid and organics

1914 seems to be enhanced by the surface irregularities in the soot. The typical fractal agglomerate
 1915 structure of soot may offer a large specific surface area for adsorption and chemical reactions
 1916 (Popovitcheva et al., 2000). Recently, Loukhovitskaya et al. (2013) also investigated the uptake of
 1917 HNO_3 on aviation soot.
 1918
 1919 The EIs of elemental and organic carbon were investigated during APEX campaigns (Kinsey, 2009;
 1920 Onasch et al., 2009): results showed that EC ranged from 21 to 98 mg kg Fuel⁻¹ and OC between 37
 1921 and 83 mg kg Fuel⁻¹. Most studies indicated that BC emissions are a function of engine thrust
 1922 settings (Anderson et al., 2005; Wey et al., 2007; Kinsey, 2009; Kinsey et al., 2011), but are nearly
 1923 independent of FSC (e.g., Wilson et al., 2004; Kinsey, 2009). During the EXCAVATE campaign,
 1924 Anderson et al. (2005) concluded that black carbon emission indices increase significantly from idle
 1925 to cruise power. These findings are also consistent with the results of the APEX campaigns: Wey et
 1926 al. (2007) and Kinsey et al. (2011) reported that BC emissions are minimum at low power and
 1927 increase with thrust settings, reaching values more than 0.3 g kg Fuel⁻¹ at power levels higher than
 1928 85% F_{00} and dominating the total mass emissions. Agrawal et al. (2008) reported that the
 1929 carbonaceous PM composition (EC+OC mass) significantly increases with power and shifts from
 1930 OC-rich at idle to EC-rich with rising thrust regimes. Similar findings were observed by Petzold
 1931 and Schröder (1998), who indicated that the ratio of BC to total carbon ranged from 11% at idle to
 1932 >80% at take-off thrust. This result is predictable when considering that the highest emissions of
 1933 hydrocarbons occurs at low power. Presto et al. (2011) recently investigated both the elemental
 1934 carbon and the organic aerosol emitted by a CFM56-series engine at varying thrust settings after the
 1935 exhaust using a smog chamber. Their findings confirmed the U-shaped curves of PM emissions
 1936 versus thrust commonly reported in the literature, but also added new important knowledge on the
 1937 relative contributes of EC and OA. At low power (4%–7% F_{00}), most PM is composed of OA, while
 1938 at 30% thrust very low emissions of both elemental and organic components were observed. At

1939 climb power (85%), an abrupt increase of EI(PM) occurred, mainly driven by EC, which accounted
1940 for about two thirds of the total PM.

1941

1942 The chemical characterisation of the organic component of the PM indicated that over 70% of the
1943 particle-phase organic compounds are made up of SVOC compounds in the *n*-alkane (mainly C₂₃ to
1944 C₃₃), PAH, and sterane/hopane compound classes (Kinsey et al., 2011). Besides the lighter PAHs,
1945 which mainly partition in the gaseous phase, the heavier congeners are principally in the particulate
1946 phase and generally also have the highest carcinogenic and mutagenic potencies (Delgado-Saborit
1947 et al., 2011). Hu et al. (2009) studied the effect of aircraft movements at a site located 100 m
1948 downwind of the regional airport of Santa Monica and reported spikes in concentration of particle-
1949 bound PAHs occurring during jet take-offs (440 ng m⁻³, i.e. 90 times the local background levels),
1950 however they did not detect significantly higher average levels of PAHs at airports. It is interesting
1951 to note that PAH emissions at airports may also undergo local deposition. In a study carried out at
1952 Delhi International Airport, Ray et al. (2008) observed that PAH contamination in the <2 mm
1953 surface soil layer reached maximum levels at a site near the landing area. The presence of PM-
1954 bound hopanes and steranes is also intriguing because these compounds are present in crude oil and
1955 are also largely used as molecular markers of vehicle emissions (e.g., Zielinska et al., 2004; Kam et
1956 al., 2012). Additional insights are therefore necessary for the characterisation of these organic
1957 compounds, which can derive either from the unburned fuel or from the emission of lubricating oils,
1958 which was hypothesised to have an important role in the mass of organic PM (Yu et al., 2010).

1959

1960 The emission of carbonaceous PM was also reported in further studies conducted at airports. For
1961 example, Dodson et al. (2009) performed continuous BC measurements at five monitoring sites in
1962 close proximity to a small regional airport in Warwick, Rhode Island. By coupling BC data with
1963 real-time flight activities (departures and arrivals) and meteorological data, they reported that
1964 aircraft departures and arrivals (and other sources coincident in space and time) contribute

1965 approximately 24-28% of the total BC concentrations. Further, they also indicated that aircraft take-
1966 off makes a greater contribution to BC levels than landing. Hu et al. (2009) studied the effect of
1967 aircraft movements in a neighbourhood adjacent to the regional airport of Santa Monica and
1968 generally did not observe elevated average levels of BC, although spikes in concentration of this
1969 pollutant were observed associated with jet take-offs. At a site located 100 m downwind of the take-
1970 off area, jet departures resulted in short time (60 s) peaks with average concentrations of up to 30
1971 $\mu\text{g m}^{-3}$, i.e. 100 times elevated above the local background.

1972

1973 **4.14.2 The smoke number (SN)**

1974 Despite soot corresponding to the majority of the non-volatile mass of PM emitted by aircraft, this
1975 component is not directly certified by ICAO. However, the ICAO databank requires that an exhaust
1976 opacity metric called the smoke number (SN) is measured for TF engines. SN was defined as a
1977 “dimensionless term quantifying smoke emission level based upon the staining of a filter by the
1978 reference mass of exhaust gas sample and rated on a scale of 0 to 100” (ICAO, 2008). SN was
1979 firstly collected on a filter by flowing a defined volume of the exhaust gas (12 to 21 kg of exhaust
1980 gas per square meter of filter) by a sample probe positioned directly behind the engine nozzle and
1981 inside the exhaust jet. The degree of attenuation of the filter before and after the sampling was thus
1982 measured using a reflectometer, and the SN was computed as:

$$1983 \quad \text{SN} = 100 \cdot (1 - R_f / R_0)$$

1984 where R_0 and R_f are the absolute reflectance of the filter before and after the sampling, respectively.
1985 Unfortunately, SN gives only a qualitative estimate of particle emission and was recognised to be
1986 dependent on sampling conditions, soot characteristics and morphology, and therefore was assumed
1987 to have little value for estimating atmospheric impacts (Anderson et al., 2005). Moreover, it was
1988 reported that particles with a diameter less than 300 nm passed through the filter and therefore only
1989 the larger particles are collected resulting in a relative weak accuracy of measurement (Kugele et
1990 al., 2005).

1991 Several studies have attempted to correlate SN to BC mass concentration (e.g., Champagne, 1971;
1992 Whyte, 1982; Girling et al., 1990; Petzold and Döpelheuer, 1998; Wayson et al., 2009; Peck et al.,
1993 2013; Stettler et al., 2013a,b) and today an interim methodology named first-order approximation
1994 3.0 (FOA3) was developed and used to estimate BC mass emissions normalised by fuel burn
1995 EI(BC) from SN (Wayson et al., 2009). Although this calculation was reported to be dependent
1996 upon the mode-specific SN recorded in the engine databank (e.g., Stettler et al., 2011), recently
1997 Stettler et al. (2013b) observed that the correlation between BC and SN depends on the particle size
1998 distribution and that the methods suggested to convert SN to BC could lead to heavy
1999 underestimations of BC concentrations. An alternative method independent of the SN (FOX) was
2000 also recently developed and first studies reported an improved estimation of BC (Stettler et al.,
2001 2013a), but it needs to be further tested. To fill this gap, recently an group of experts was called to
2002 define new standard procedures for BC measurement at ground level for regulatory purposes (SAE,
2003 2009). In the absence of defined standards, the scientific literature offers a number of studies on the
2004 emission of soot, BC and EC.

2005

2006 **4.14.3 Inorganic ions**

2007 The analysis of the major inorganic ions in aircraft exhaust has a clear dependence on the adopted
2008 sampling methodology and can be affected by many artefacts. As for most hydrocarbons, ions may
2009 undergo gas-to-particle partitioning and some species may further derive from chemical reactions in
2010 the atmosphere or on the filter surface. For example, the concentrations of aerosol nitrate can be
2011 affected by the adsorption of nitric acid gas on pre-existing particles, while evaporative losses occur
2012 at temperatures $>20\text{ }^{\circ}\text{C}$ and the exhaust plumes largely exceed this temperature. In addition,
2013 sulphate may form quickly due to the oxidation of SO_2 , coating soot particles. In view of this,
2014 Anderson et al. (2005) firstly reported that the concentration of sulphate aerosol rose considerably
2015 as sampling was performed progressively downstream of the engine, suggesting that sulphate
2016 particles may originate or undergo rapid growth within aircraft exhaust plumes. These findings were

further confirmed by APEX campaigns. Agrawal et al. (2008) noted that the mass of the ions collected at 1 m from the engine exit plane were below the detection limit for most ions, while only sulphate was detectable. On the contrary, APEX samplings at 30 m reported EI(ions) in the range of 30-40 mg kg Fuel⁻¹ dominated by sulphate (53%–72% of the total ion EIs) and ammonium (Kinsey et al., 2011). In summary, there is a lack of data on the ionic component of exhaust emissions of aircraft and this merits further investigation.

2023

4.14.10 *Elemental composition*

There is a severe shortage of data on the elemental composition of PM emitted by aircraft. Kinsey et al. (2011) reported that PM_{2.5} emissions are composed of various trace elements mainly originating from fuels, lubricating oils, engine wear and corrosion, although release from the sampling line and fugitive dust may contribute to the total load. During the APEX campaigns, the elemental composition of PM emitted from aircraft engines was analyzed for a number of different aircraft engines. The total elemental emissions (sum of Mg, Si, P, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Br, Ag, In, Sb, Te, I, Tl) were in the range of 6.3—27.5 mg elements kg Fuel⁻¹, corresponding to 2–7% of the total emitted PM and were dominated by sulfur (54%-80% of total element mass) (Kinsey, 2009; Kinsey et al., 2011). As expected, sulfur was well correlated with sulphate and most of the sulfur on the filter exists as sulphate (Agrawal et al., 2008). Moreover, the variability in the metal emissions was observed to be much greater between different engines than between engine thrust settings (Agrawal et al., 2008).

2037

Recently, Mazaheri et al. (2013) investigated the physical and chemical characteristics of individual particles collected in the exhausts of in-use aircraft during landing and takeoff by using transmission microscopy and energy dispersive X-ray spectroscopy. They reported that most of the measured particles have a spherical shape in the nucleation mode (18–20 nm) and only contain C, O, S, Cl, and in some cases K. They also reported fewer particles having a more irregular shape

2043 resulting in a larger average aspect ratio and a much greater and diverse range of elements. While
2044 the small spherical particles have been linked to the combustion processes of engines, the latter
2045 irregular particles have been linked to a diverse range of sources, including tyre wear, fine dusts,
2046 vehicular traffic, and possibly engine wear.

2047

2048 ***4.14.12 Secondary aerosol***

2049 Despite the potential role of aircraft emissions in forming SIA and SOA, there is a lack of
2050 information on the chain of processes affecting aircraft emissions once emitted in ambient air. A
2051 recent study by Miracolo et al. (2011) used a smog chamber to simulate the aging of the particulate
2052 matter emitted from a TF engine under typical (summertime) atmospheric conditions. Their
2053 findings pointed out the key role of the photo-oxidation processes in forming both SIA and SOA.
2054 They reported that after several hours of photo-oxidation, the ratio of secondary-to primary PM
2055 mass was on average 35 ± 4.1 , 17 ± 2.5 , 60 ± 2.2 and 2.7 ± 1.1 for increasing thrusts settings (4%, 7%,
2056 30% and 85% F_{00} , respectively). Miracolo et al. (2011) also observed that SOA dominates the
2057 secondary PM at low thrust, while secondary sulphate becomes the main secondary component at
2058 higher power.

2059

2060 It is not clear if aircraft emissions can influence the amount of secondary aerosol on a large scale. In
2061 this regard, a recent study by Woody and Arunchalam (2013) used the Community Multiscale Air
2062 Quality (CMAQ) model to investigate the impacts of aircraft emissions on SOA at the Hartsfield-
2063 Jackson Atlanta International Airport. By applying the model at various spatial resolutions, they
2064 reported that aircraft emissions reduced SOA by ~6% at 36 and 12-km due to the chemistry of the
2065 free radicals with aircraft NO_x , while at smaller resolution the interaction between the aircraft
2066 emissions and external biogenic SOA precursors enhanced SOA (~12%).

2067

2068

2069 **5. AIRCRAFT NON-EXHAUST EMISSIONS**

2070 Although the vast majority of studies have focussed upon the exhaust emissions from engines, there
2071 are other aircraft-related emissions that may influence the air quality within an airport. These
2072 include emissions from the power units, i.e. APU's and GPU's, primary particles from tyre erosion
2073 and brake wear, oil leaks and corrosion of aluminium alloys, all of which have been recognised to
2074 impact air quality near airports but at date have received only limited consideration.

2075

2076 **5.1 Tyre, Brake and Runway Surface Wear**

2077 Tyre and brake wear during landing and runway dust re-suspension have been estimated to be major
2078 sources of particulate matter. This is expected as smoke is clearly visible to the naked eye when
2079 aircraft wheels contact the ground and spin up to the landing velocity. Despite that, the proportion
2080 of the mass lost from aircraft tyres and brakes that becomes suspended as fine PM has not been
2081 extensively studied; the few available data indicate that the rubber lost from tyre wear can vary
2082 from few grams to ~0.8 kg per landing (Morris, 2006; Bennett et al., 2011 and references therein).
2083 Particulate emissions from tyres have been suggested to be dependent upon the maximum take-off
2084 weight, but other factors may have a role in the rubber wear, e.g., number of wheels, weather
2085 conditions, engine type, airport runway length and taxiway layout and operating procedures
2086 (Morris, 2006). The subsequent activation of brakes to bring the aircraft to a stop may further
2087 abrade brake lining material from discs and pads and may release fine particles as for road vehicles
2088 (e.g., Pant and Harrison, 2013). From a physicochemical point of view, it is plausible that brake
2089 wear includes both the emission of material from the abrasion of discs and the volatilisation and
2090 condensation of brake pad materials, while soot may arise from the thermal degradation of tyre
2091 polymers. This was confirmed by experimental data collected at a major European airport: Amato et
2092 al. (2010) reported unusually high levels of both organic carbon and metals possibly sourced from
2093 tyre detritus/smoke in runway dust (Ba, Zn, Mo) and from brake dust in ambient PM₁₀ (Cu, Sb). In

2094 addition to tyre and brake wear, landing field wear and re-suspension can also occur, as usually
2095 aircraft land on a runway generally constructed of asphalt, concrete, gravel or grass.

2096

2097 For example, studies at Gatwick airport estimated that tyre and brake wear are dominant sources of
2098 PM_{10} , accounting about 22 and 4.5 tonnes y^{-1} , respectively, i.e. about 60% and 12% of all aircraft-
2099 related emissions, respectively (British Airports Authority, 2006). However, these emissions are
2100 subject to large uncertainties as they are dependent on many factors, including speed at landing,
2101 some aircraft characteristics (weight, number of wheels, brake material if carbon or steel) and
2102 runway characteristics (length, weather conditions) (Underwood et al., 2004).

2103

2104 Bennett et al. (2011) collected landing and braking dust samples from the undercarriage (oleo legs)
2105 and wheel hubs of aircraft and reported that they have bimodal distributions, with peaks at
2106 aerodynamic diameters of about 10 and 50 μm . A further SEM-EDS analysis has revealed that
2107 particles may contain various materials embedded in a carbonaceous substrate: (i) soot arising from
2108 the burning of the tyre rubber, from the asphalt tar or from brake abrasion; (ii) runway dust mainly
2109 composed of typical crustal materials (quartz and feldspar particles) which are lifted mechanically
2110 from the ground surface; (iii) small droplet (35 μm) of Fe, associated with Co and other transition
2111 metals (Mn, Ni, V, Zn) which are commonly found in asphalt concrete and (iv) irregular Fe
2112 particles (<10 μm). This study also reported that aluminium, which is typically used as tracer for
2113 crustal materials from runway wear, can also derive from Al hydroxide included in some tyre
2114 formulations.

2115

2116 **5.2 Other Mechanical Components**

2117 High-strength aluminium alloys are commonly used as the aircraft fuselage materials in the body
2118 and wings, while minor amounts of other elements (Cu, Zn, Mg) may be also present in various
2119 airframe components (Wei et al., 1998). Aluminium alloys have a microstructure that can be highly

susceptible to intergranular and pitting corrosion, and weathering is recognised as a major cause of structural damage to aircraft structure and coatings (Usmani and Donley, 2002; Russo et al., 2009; Knight et al., 2011), along with long term operations (Ostash et al., 2006), runway de-icing chemicals (Huttunen-Saarivirta et al., 2011) and atmospheric pollution and salts (Cole and Paterson, 2009). The degradation of aircraft mechanical components is also connected with mechanical, and corrosion-mechanical (macrocracks) defects, which lead to a decrease in its load-bearing capacity (Ostash et al., 2006). Corrosion has many forms and affects most structural alloys found in airframes: of particular importance is pitting and intergranular corrosion, which can develop into fatigue cracks, stress corrosion cracks or exfoliation (Liao et al., 2008). In this light, it is plausible that corrosion and mechanical stress of some aircraft components may release metallic particles into the environment. For example, using scanning electron microscopy techniques, Amato et al. (2010) founded the relatively common presence of platy aluminous particles derived from airframe corrosion in the ambient PM₁₀ samples collected near the El Prat airport in Barcelona.

2133

2134 **5.3 Oil Leaks**

In addition to exhaust from jet fuel combustion, oil escaping or burning from lubricated parts may be vented overboard from aircraft engines and therefore may further contribute to the total emissions of aircraft (Onash et al., 2009; Timko et al., 2010b; Yu et al., 2010; 2012). Aircraft lubricating oils are usually composed of a mixture of synthetic C₅-C₁₀ fatty acid esters of pentaerythritol and dipentaerythritol with specialised additives (Yu et al., 2010; 2012). Some of these, such as tricresyl phosphate, are recognised as toxic to humans (Craig and Barth, 1999; Van Netten, 1999; Winder and Balouet, 2002; Marsillach et al., 2011) and have been detected in ambient air and aircraft cabins, posing a risk for aviation technicians, loaders, crew and passengers in case of release into the environment (e.g., Solbu et al., 2010; Liyasova et al., 2011; Denola et al., 2011; Schindler et al., 2013). Yu et al. (2010) reported that the degree of degradation of lubrication oil during aircraft engine operations as a result of friction and/or pyrolysis might be negligible,

2146 suggesting that most emitted oil is unburned. Because of its low volatility, unburned lubricating oil
2147 may exit from engines as vapour or submicrometre droplets and may further condense and add mass
2148 to the organic PM in the wake of the aircraft. Results of exhaust characterisation measurements
2149 suggest that the contribution of lubrication system releases to the organic PM may be greater than
2150 the engine exhaust (Timko et al., 2010b): they estimated that the contribution of oil leaks to the total
2151 mass of organics generally lies within the range 10-20% for low thrust and 50% for high thrust
2152 settings. A recent study (Yu et al., 2012) has identified and quantified the lubricating oil in the
2153 particulate matter emissions from various engines of in-service commercial aircraft at two airports.
2154 This study used the characteristic mass marker of lubricating oil (ion fragment intensity between
2155 $m/z = 85$ and 71) to distinguish lubricating oil from jet engine combustion products. Results
2156 revealed that lubricating oil is commonly present in organic PM emissions in association with
2157 emitted soot particles, unlike the purely oil droplets observed at the lubrication system vent. The
2158 contribution from lubricating oil in aircraft plumes was observed to vary from 5% to 100% in
2159 measured aircraft plumes.

2160

2161 Yu et al. (2010) measured the size distributions of submicrometre unburned lubricant oil released
2162 from engines with C-TOF-AMS and UHSAS and reported a shift to larger sizes with increasing
2163 power. At idle thrust they observed a C-TOF-AMS vacuum aerodynamic diameter (D_{va}) of 260 ± 3
2164 nm, while the UHSAS volume equivalent diameter (D_{ve}) was 281 ± 9 nm. At higher engine power,
2165 they observed modes at 272 ± 4 nm and 350 ± 8 nm for C-TOF-AMS and UHSAS, respectively.

2166

2167 **6. OTHER AIRPORT-RELATED EMISSIONS**

2168 Apart from aircraft exhaust and non-exhaust emissions, other sources can be present within an
2169 airport and can contribute to the total pollutant load in the atmosphere. Among others, the emissions
2170 of the power units providing power to the aircraft (APUs and GPUs), the GSEs, additional sources

2171 on the modern terminals, intermodal transportation systems and road traffic are further considered
2172 as impacting upon the air quality and must be taken in account in airport emission measurements.

2173

2174 **6.1 Auxiliary and Ground Power Units**

2175 The APU's are small on-board gas-turbine engines burning jet fuel coupled with an electrical
2176 generator capable of supplying electrical power to aircraft systems when required on the ground or
2177 providing pneumatic or hydraulic power to start the main engines. Despite APU's being installed in
2178 all modern airliners so as to be energetically independent, their use is becoming less significant over
2179 time due to the increasing trend toward mains supplied Ground Power Units (GPU) (Mazaheri et
2180 al., 2011). This ground equipment is supplied by the airports and includes diesel powered tugs of
2181 various types, ground carts, and also APU's installed on ground carts (e.g., Kinsey et al., 2012b).
2182 Some airports also provide electrical power to the aircraft by connecting directly to the ground
2183 network and by using fixed ground electrical power (FGEP) units. This system avoids the use of
2184 fuelled power units, with a subsequent reduction in local emissions and is thus very useful in
2185 airports not complying with air quality standards.

2186

2187 The role of the APU's on the air quality at airports is nowadays widely discussed and an increasing
2188 number of studies have estimated their contribution. However, the results are often conflicting.
2189 Schäfer et al. (2003) indicated that APU emissions at airport service buildings cannot be neglected
2190 in comparison to the main engine emissions. The emission inventory of the airport of Zurich in
2191 2004 (Fleuti and Hofmann, 2005) reported that although the aircraft exhaust accounted for most of
2192 CO, hydrocarbons and NO_x (89%, 45%, 82%, respectively of total emissions), a significant percent
2193 was from APU's, GPU's, start-up-idle, handling/GSE, airside traffic and stationary sources, with
2194 APU's accounting for about half of the total non-aircraft engine emissions. HAL (2011) reported
2195 that 19% of the total NO_x emissions of London Heathrow airport are due to the use of APU's. A
2196 survey over 325 airports in the USA (Ratliff et al., 2009) estimated the emissions from APU's and

2197 LTO cycles and stated that the greatest percentage that APUs contributed to total aircraft emissions
2198 was 10-15% for CO and between 15 and 30% for NO_x and SO_x. However, this study also reported
2199 that the airports used by a higher percentage of small and business jets tend to be affected by higher
2200 emissions from the APUs. Stettler et al. (2011) estimated that APUs contribute 6% to total PM_{2.5}
2201 emissions at major UK airports. The effect of the APUs upon public health was recently estimated
2202 by Yim et al. (2013), who calculated the emissions from aircraft LTO activity, aircraft APUs and
2203 GSE at the top 20 UK airports, ranked by passenger numbers. Their findings concluded that the ban
2204 on the use of APUs would prevent about 11 averted early deaths per year (90% confidence interval
2205 7-16).

2206

2207 Unlike aircraft engines, APU emissions are not certificated by ICAO, and the manufacturers
2208 generally consider information on APU emissions rates as proprietary (ICAO, 2011), therefore there
2209 are today few data available on APU emissions. Emissions from APU depend on many factors and
2210 are subject to change through provision of GPU facilities from the airport. Some airports have
2211 implemented policies to encourage the use of the GPU instead of APUs (Mazaheri et al., 2011 and
2212 reference therein), however in the absence of GPU availability, the use of APUs is still the only
2213 alternative to provide the energy for aircraft operations with engines off and for the ignition of the
2214 engines. The first studies of APU emissions started in the 1970s by the US Army (Kinsey et al.,
2215 2012b and references therein) and our literature search has found very few data in comparison to
2216 those on the jet engine emissions. However, the main studies reporting (or reprocessing) data on the
2217 APU emissions are increasing nowadays (Slogar and Holder, 1976; Williams and Lee, 1985;
2218 Gerstle et al., 1999; 2002; Wade, 2002; O'Brien and Wade, 2003; Schäfer et al., 2003; Watterson et
2219 al., 2004; EASA, 2011; Anderson et al., 2011; Blakey et al., 2011; Kinsey et al., 2012b; Williams et
2220 al., 2012).

2221

2222

2223 **6.2 Ground Service Equipment Emissions, Vehicular Traffic and Other Sources**

2224 As they are strictly linked to the airport operations, the amount of GSE vehicles clearly reflects the
2225 airport layout and traffic in terms of both cargo and passengers. Moreover, the operation duration is
2226 expected to increase with increasing aircraft size. Other factors include the type of engines installed
2227 and the quality of fuels used and the status of the vehicle fleet (age, wear and tear). Therefore, it is
2228 not possible to identify the unique characteristics common to all the airports and ICAO databanks
2229 not include any information about GSE emissions. Similarly, the amount of road traffic in the form
2230 of private cars, taxis, shuttle bus and trucks for transporting people and goods in and out to the
2231 airport depends on the airport layout, on the quality of the road links and intermodal transport
2232 systems and, finally, is directly related to the number of passengers and goods that the airport
2233 handles. As both the airport-induced vehicular traffic and most of the GSEs have gasoline or diesel
2234 engines, it is reasonable to consider them as common traffic. The traffic source is recognised to be
2235 dominant in many urban environments. Its chemical and physical characteristics are reported
2236 elsewhere, in a large number of studies and reviews (e.g., Hueglin et al., 2006; Thorpe and
2237 Harrison, 2008; Johansson et al., 2009; Gietl et al., 2010; Kumar et al., 2011; Harrison et al., 2012;
2238 Pant and Harrison, 2013; Amato et al., 2013).

2239
2240 Some studies have indicated that GSE may contribute a major fraction of the total AEs. For
2241 example, a study carried out at the McCarran airport in Las Vegas reported that approximately 60%
2242 of the total airport emissions are related to GSE (Nambisan et al., 2000). Schürmann et al. (2007)
2243 calculated that NO concentrations at Zurich airport were dominated by emissions from ground
2244 support vehicles, while Unal et al, (2005) estimated that the impacts on ozone and PM_{2.5} of GSE at
2245 the Hartsfield–Jackson Atlanta International airport are small compared to the aircraft impacts. In
2246 addition, other miscellaneous sources may be also present at airports and may further increase the
2247 total pollutant load, including maintenance work, heating facilities, fugitive vapours from refuelling
2248 operations, kitchens and restaurants for passengers and operators, etc. Despite being intermittent

2249 and depending on the airport layout, these emissions may be dominant in certain circumstances. For
2250 example, Amato et al. (2010) reported that the local construction work for a new airport terminal in
2251 a major European airport (El Prat, Barcelona) was an important contributor to PM₁₀ crustal dust
2252 levels along with road dust and aircraft re-suspension, with a clear drop during the weekends.

2253

2254 7. AIRPORT EMISSIONS AND PUBLIC HEALTH

2255 While aircraft emissions at cruising altitudes are an air pollution issue at global scale (Barrett et al.,
2256 2010; Koo et al., 2013), the emissions within the planetary boundary layer due to the LTO
2257 operations are certainly more local and it is plausible to believe they may have a more direct effect
2258 on human health. Nevertheless, the potential subsidence of air masses due to the Ferrell and Hadley
2259 circulations, which may displace high altitude emissions toward the ground cannot be disregarded
2260 (Barrett et al., 2010).

2261

2262 Air quality degradation in the locality of airports is considered by some to pose a real public health
2263 hazard (Barrett et al., 2013) and some recent estimates of the aviation contribution to premature
2264 mortality have been reported (e.g., Ratliff et al., 2009; Levy et al., 2012; Ashok et al., 2013, Yim et
2265 al., 2013). Although at the current time, no specific target toxic compound has been identified to be
2266 used as a marker or indicator for human exposure to jet engine fuels and their combustion products
2267 (Tesseraux, 2004), it has been estimated that over 2 million civilian and military personnel per year
2268 are occupationally exposed to jet fuels and exhaust gases (Pleil et al., 2000; Ritchie, 2003; Cavallo
2269 et al., 2006). Kerosene-based fuels have the potential to cause acute or persistent neurotoxic effects
2270 from acute, sub-chronic, or chronic exposure of humans or animals (Ritchie et al., 2001), although
2271 evidence is lacking that current levels of exposure are harmful. Occupational exposure can occur by
2272 dermal, respiratory or oral ingestion routes of raw fuel, vapour, aerosol or exhausts. It has been
2273 postulated that chronic exposure to vapours and exhaust fumes could affect the operators inside the
2274 airport (Cavallo et al., 2006) and aircraft crew (Denola et al., 2011; Schindler et al., 2013), while

occasional exposure can affect all passengers in transit (Liyasova et al., 2011). In addition, also the population living in the vicinity of airports can be exposed (Jung et al., 2011).

However, the impact of LTO emissions on surface air quality and human health is poorly quantified (Barrett et al., 2010) even though most governments have recently focused attention on management and reduction the environmental impacts of aviation. Some studies have attempted to estimate the direct and indirect effects of aviation to support environmental policy assessments and to evaluate many possible future scenarios. A global-scale study by Barrett et al. (2010) estimated that ~8000 premature deaths per year can be attributed to aircraft emissions at cruising altitudes, representing ~80% of the total impact of aviation (including LTO emissions) and ~1% of air quality-related premature mortalities from all sources.

A series of more local studies have been conducted to assess the impact of AEs on human health. Generally the results have highlighted the potential adverse effects of AEs on public health and also revealed the need for more extensive information about this source. Three estimates were given for US airports in 2005: Ratliff et al. (2009) analysed aircraft LTO emissions at 325 US airports with commercial activity and estimated that 160 (90% confidence interval 64-270) premature deaths occurred due to ambient particulate matter exposure attributable to the aircraft emissions; Levy et al. (2012) estimated about 75 early deaths using activity data from 99 US airports; Ashok et al. (2013) estimated that aviation LTO emissions caused about 195 (90% confidence interval 80-340) early deaths, while the same emissions were forecast to cause ~350 (90% confidence interval 145-610) deaths in 2018. Arunachalam et al., (2011) used the Community Multiscale Air Quality model (CMAQ) to estimate the incremental contribution to PM_{2.5} due to commercial aviation emissions during LTO cycles in two major and one mid-sized US airport and reported that 8-9, 11-15 and 5 (depending on model resolution) premature deaths per year can be estimated for Atlanta, Chicago and Providence airports, respectively. In Europe, Yim et al. (2013) estimated that 110 (90% CI:72-

160) early deaths occur in the UK each year (based on 2005 data) due to airport emissions. The same study also assessed that up to 65% of the health impacts of UK airports could be mitigated by replacing current fuel with low FSC fuel, by electrifying GSE, avoiding use of APUs and use of a single engine during the taxi phase. Lin et al. (2008) estimated that residents living within five miles of Rochester and La Guardia airports are affected by an increased relative risk of hospital admission of 1.47 and 1.38 respectively compared to resident living >5 miles distant. Jung et al. (2011) characterised the levels of BTEX in the vicinity of the Teterboro airport, New York/New Jersey metropolitan area, by exposing passive samplers for 48 h at the end of airport runways, in households close to the airport and out-of-neighbourhood locations. Results indicated that the average concentrations of benzene, toluene, ethylbenzene, m-/p-xylenes and o-xylene in neighbourhood concentrations (0.8, 3.8, 0.4, 1.2 and 0.4 $\mu\text{g m}^{-3}$, each BTEX respectively) were not significantly different to those measured at the airport runways (0.8, 3.2, 0.3, 1, and 0.3 $\mu\text{g m}^{-3}$, respectively) and higher than the out-of-neighbourhood locations (0.5, 1.1, 0.2, 0.8, and 0.4 $\mu\text{g m}^{-3}$, respectively). Cavallo et al. (2006) characterised the exposure to PAHs in airport personnel and evaluated the genotoxic and oxidative effects in comparison with a selected control group. They analysed 23 PAHs collected from various areas over five working days and urinary 1-hydroxypyrene (1-OHP) following five working days as a biomarker of exposure. They reported an induction of sister chromatid exchange due to PAH exposure, although its health significance was not quantified.

2320

2321 **8. CONCLUSIONS**

2322 The main goal of this review is to give an overview on the current state of knowledge of airport-
2323 related emissions and to summarise the key characteristics of pollution and the impacts on local and
2324 global air quality. After thoroughly reviewing the latest available scientific literature, it can be
2325 concluded that the currently available information on the impact of AEs upon air quality is
2326 inadequate and the consequences of future growth in the volume of air traffic are very hard to

2327 predict. Most work has focussed upon aircraft engine exhaust during LTO cycles which accounts
2328 for a large proportion of the total emitted pollutants. However other sources such as the auxiliary
2329 power units, vehicular traffic and ground service equipment are known sources that may seriously
2330 affect air quality near to airports. In this way, it is apparent from the literature that while aircraft
2331 exhaust may account for most of the pollution at some airports, there are other sources that need to
2332 be addressed in more detail in the future, such as:

2333

- 2334 • tyre, brake, asphalt wear and the re-suspension of particles due to the turbulence created by
2335 aircraft movements;
- 2336 • the emissions from the units providing power to the aircraft when required on the ground
2337 (APUs and GPUs);
- 2338 • the ground support equipment that an airport offers as a service for flights and passengers,
2339 including passenger buses, baggage and food carts, container loaders, refilling trucks,
2340 cleaning, lavatory servicing and de/anti-icing vehicles, and tugs;
- 2341 • the effects of the intermodal transportation systems, and road traffic for transporting people
2342 and goods in and out to the airport.

2343

2344 Most studies report that airport operations are responsible for significant emissions of a series of
2345 non-volatile, gaseous and semi-volatile species. Non-volatile emissions are made up of refractory
2346 material such as soot, which is emitted as PM even at high temperatures, but is also comprised of
2347 many organics and sulfur compounds, the latter mainly in the form of sulphate. Volatile emissions
2348 include compounds that exist as vapour at the engine exit plane and are made up of gaseous and
2349 vapour-phase pollutants, such as CO, NO_x, SO₂ and many organics (i.e. aromatics, alkanes, alkenes
2350 and a number of other VOCs). The less volatile fraction is of especial interest as it can react in the
2351 atmosphere and undergo gas-to-particle conversion by forming new particles or condensing on pre-
2352 existing ones.

2353

2354 The volatile emissions have mostly been fairly well characterised, but a comprehensive chemical
2355 speciation of the hydrocarbons and complete knowledge of their chemical processing in the
2356 atmosphere is still lacking. Detailed information on the non-volatile and semi-volatile compounds is
2357 also scarce. In spite of the increasing attention given to AEs, many issues remain unaddressed and
2358 represent a serious gap on which scientific research should focus. A list of the key characteristics
2359 of AEs that need to be carefully addressed should include:

2360

- 2361 • a careful quantification of sulfuric acid, HONO and HNO₃ directly emitted by aircraft for a
2362 large variety of engines. Currently available data refer only to few engine types and the
2363 changes of EI at varying thrusts are not completely clear. This should also include seeking a
2364 better knowledge of the characteristics and the evolution of emitted chemi-ions and a better
2365 understanding of their role as a source of sulfur and nitrogen species in plumes;
- 2366 • a more realistic quantification of emission inventories for nitrogen oxides and organic
2367 compounds, which includes the variability induced by the common practices of take-off and
2368 taxi phases at reduced thrust;
- 2369 • quantification of the effects of ozone-precursors emitted from aircraft and other AEs on the
2370 levels of ground-level ozone at airports, which to date have not been thoroughly investigated.
2371 In particular, since well established atmospheric photochemical reactions of many VOCs are
2372 known as potential sources of elevated ozone concentrations in the troposphere, improved
2373 chemical speciation of organic compounds is much needed. Better apportionment of ozone
2374 formation potential from aircraft emissions during LTO cycles and from other AEs should be
2375 also estimated;
- 2376 • standardization of procedures for measurement of engine exhaust at ground level for
2377 regulatory purposes, which appear to be lacking mainly for PM and speciated hydrocarbon
2378 emissions. Such methodologies should take into account the semi-volatile components, which

2379 have been recognised to make a major contribution to the total mass of emitted PM.

2380 Achievement of this objective is vital to be able to obtain data that are comparable across

2381 different studies;

2382 • further quantitative knowledge of the chemical and physical modifications affecting many

2383 compounds and particulate matter in the atmosphere, including the oxidation of hydrocarbons

2384 to less volatile species and the formation of sulphate on the surface of pre-existing particles;

2385 • chemical and physical characterization of PM. Far fewer data exist for PM than for the main

2386 gaseous pollutants. The chemical speciation of PM is not fully understood and the role of

2387 plumes aging on PM mass and composition is largely unknown. The role of lubrication oils,

2388 fuel type and engine technology, age and maintenance upon aircraft PM emissions also needs

2389 to be investigated;

2390 • a more detailed assessment of the health effects of the AEs within and in the surroundings of

2391 major airports;

2392 • the identification of particular chemical species to be used as tracers for most of the AE

2393 sources;

2394 • the significance of airport operations for emission reduction and management should be

2395 investigated in more depth. There is a lack of information on the effects of time-in-modes,

2396 aircraft waiting/idling durations, aircraft weight, and use of APU/GPU/FGEP on the actual

2397 emission of pollutants. A more detailed knowledge of such operations will lead to a more

2398 reliable assessment of the quantities of exhaust pollutants emitted into the air;

2399 • the relative importance of near-airport, regional, and global scale air quality impacts of airport

2400 and aircraft emissions need to be further investigated. Most studies focus on local or global

2401 effects of the AEs, but there is no comprehensive view of air pollution over a full range of

2402 scales.

2403

2404 Quantification of the impact of airport emissions on local air quality is very difficult due to the
2405 complexity of airport emissions and the presence of substantial levels of pollution from other
2406 sources, with many airports being located near to urban settlements, major highways and roads or
2407 industrial installations. This makes the signal of the AEs and, in particular, of aircraft emissions
2408 very hard to distinguish. This is a serious gap because development of cost-effective strategies to
2409 improve air quality to meet regulatory requirements demands a clear quantification of the
2410 contribution of AEs to the total air pollution.

2411

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3993 **TABLE LEGENDS**

3994 **Table 1:** Engine-family mounted in the most popular aircraft. The number of engines for each
3995 aircraft in given within brackets. This list represents ~75% of total in-use turbofan
3996 engines provided by the ICAO databank at August 2013 and does not report data for
3997 regional jets. Average data (mean±standard deviation) for fuel consumption and
3998 emissions per LTO cycle are also reported per each engine family.

4000 **Table 2:** Total annual fuel burned by aviation and emissions of H₂O, CO₂, NO_x, CO, HC, SO_x
4001 and soot (when available) provided by recent studies. Forecasts for 2020 and 2025 are
4002 also provided. Global emission data for 2008 and forecasts for 2025 were calculated
4003 starting from fuel data of Chèze et al. (2011) and emission indices of Lee et al. (2010).
4004 Kim et al. (2007) provided fuel burn and NO_x emission during LTO for the 2000-2005
4005 period; LTO emissions of H₂O, CO₂ and SO₂ were calculated starting from fuel data of
4006 Kim et al. (2007) and emission indices of Lee et al. (2010). Note that all emissions
4007 calculated in this review are in italics.

4009 **Table 3:** List of recent studies in the literature that measure EIs directly from engine or airplane
4010 tests. The table also reports studies on hydrocarbon profiles. Some information about
4011 tested aircraft and engine models, selected thrust and sampling methodologies and
4012 analytical techniques, type of fuel, date and location of experiments is also given.

4014 **Table 4:** List of recent studies available in the literature reporting EIs during real aircraft
4015 operation. The table also reports supplementary information (if available) about the
4016 target of the study, period and location of experiments, tested aircraft or engine models,
4017 measured pollutants, analysed LTO phases and sampling methodologies. The list of
4018 acronyms is provided in Table 3.

4020 **Table 5:** List of recent studies available in the literature conducted at airports or in their
4021 surroundings. The table also reports supplementary information (if available) about the
4022 target of the study, period and location of experiments, tested aircraft or engine models,
4023 measured pollutants, analysed LTO phases and sampling methodologies. The list of
4024 acronyms is provided in Table 3.

4026 **FIGURE LEGENDS**

4029 **Figure 1:** Absolute growth of aviation (1930–2012) recorded by ICAO in terms of RPK, RTK and
4030 aircraft kilometres. Data refers to ICAO (2013) and were taken from Airlines for
4031 America (2013).

4033 **Figure 2:** Simplified diagram of a turbofan engine (upper left); products of ideal and actual
4034 combustion in an aircraft engine (upper right); and related atmospheric processes,
4035 products, environmental effects, human health effects and sinks of emitted compounds
4036 (bottom). Adapted from Prather et al. (1999), Wuebbles et al. (2007) and Lee et al.
4037 (2009).

4039 **Figure 3:** Division of the combustion products from an aircraft engine, adapted from Lewis et al.
4040 (1999).

4042 **Figure 4:** Geographical and vertical distributions of aviation: a) column sum of global fuel burn
4043 from scheduled civil aviation in 2005, as reported by Simone et al. (2013) using AEIC

model (Stettler et al., 2011); b) annual global vertical distribution of commercial aviation fuel burn for the NASA-Boeing 1992 and 1999 (Baughcum et al., 1996a,b; Sutkus et al., 2001), QUANTIFY 2000 (Owen et al., 2010), AERO2k (Eyers et al., 2004) and AEDT 2006 (Roof et al., 2007) datasets, taken from Olsen et al. (2013).

Figure 5: Standard ICAO LTO cycle. Adapted from ICAO (2011).

Figure 6: Burned fuel and emissions for complete standardised LTO cycle. Data from ICAO databank at April 2013 (EASA, 2013). All engines certified in each period were included in the statistics, without distinction of type, manufacturer, model or technology.

Figure 7: EIs provided by the ICAO databank (EASA, 2013). All in-use engines certified from 1976 to today (April 2013) are included.

Figure 8: Fuel burned and emissions of CO, NO_x and total unburned hydrocarbons during the four LTO phases. Data were calculated from the EIs and fuel consumption provided by the ICAO databank (EASA, 2013). All in-use engines certified from 1976 to today (April 2013) were included and reprocessed as a function of LTO stages and standard times (i.e., 0.7 min for take-off, 2.2 min for climb-out, 4 min for approach and 26 min for idle).

Figure 9: Results of the APEX campaigns. Profile (mass fractions) of individual hydrocarbon species. The single compounds are ordered to show decreasing fractions.

4068 **Table 1.** Engine-family mounted in the most popular aircraft. The number of engines for each aircraft is given within brackets. This list represents ~75% of
4069 total in-use turbofan engines provided by the ICAO databank at August 2013 and does not report data for regional jets. Average data (mean±standard
4070 deviation) for fuel consumption and emissions per LTO cycle are also reported per each engine family.

Manufacturer	Engine family	Main aircraft and number of engines	Fuel and emissions per LTO cycle (kg)			
			Fuel	CO	NO _x	HC
General Electric	CF6 series	A300 (2); A310 (2); A330 (2); B747 (4); B767 (2); MD DC-10 (3); MD-11 (3)	811±76	11±5	12±2	2.3±2.2
	GE90 series	B777 (2)	1159±141	14±7	25±5	1.1±0.8
	GE9x series	B747 (4); B787 (2); replacing CF6 series	827±74	7±1	10±3	0.2±0.1
CMF International	CFM56 series	A318 (2); A319 (2); A320 (2); A321 (2); A340 (4); B737 (2); MD DC-8 (4)	419±46	6±2	5±1	0.6±0.4
Pratt & Whitney	JT8D series	B707 (4); B727 (3); B737 (2); MD DC-9 (2); MD80 (2)	477±35	5±2	4±1	1±0.9
	JT9D series	A300 (2); A310 (2); B747 (4); B767 (2); MD DC-10 (3)	842±45	19±10	13±1	7±4.8
	PW 4000 series	A300 (2); A310 (2); B747 (4); B767 (2); B777 (2); MD DC-11 (3)	966±150	8±3	17±6	1±0.8
Rolls-Royce	RB211 series	B747 (4); B757 (2); B767 (2); L1011 (3); Tu-204 (2)	852±128	15±15	15±5	7.1±11.1
	Trent series	A330 (2); A340 (4); A380 (4); B777 (2); B787 (2)	817±370	5±2	19±4	0.2±0.3
BMW Rolls-Royce	BR700 series	B717 (2)	332±32	4±1	4±1	0.1±0.1
International Aero Engines	V2500 series	A319 (2); A320 (2); A321 (2); MD-90 (2)	452±35	3±0.4	6±1	0.04±0.01
Aviadvigatel' Solov'ëv	D30 series	Tu-154 (3)	622±110	21±6	5±1	5.5±2.4

4071 B (Boeing); A (Airbus); MD (McDonnell Douglas); L (Lockheed); Tu (Tupolev).

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4079 **Table 2.** Total annual fuel burned by aviation and emissions of H₂O, CO₂, NO_x, CO, HC, SO_x and soot (when available) provided by recent studies.
4080 Forecasts for 2020 and 2025 are also provided. Global emission data for 2008 and forecasts for 2025 were calculated starting from fuel data of Chèze et al.
4081 (2011) and emission indices of Lee et al. (2010). Kim et al. (2007) provided fuel burn and NO_x emission during LTO for the 2000-2005 period; LTO
4082 emissions of H₂O, CO₂ and SO₂ were calculated starting from fuel data of Kim et al. (2007) and emission indices of Lee et al. (2010). Note that all emissions
4083 calculated in this review are in italics.

Global										
Year	Fleet ^a	Fuel	H ₂ O	CO ₂	NO _x ^b	CO	HC	SO _x ^c	Soot	Reference
Tg									Mg	
1999	Scheduled air traffic which includes turboprops, passenger jets, and jet cargo aircraft	128	—	—	1.7	0.685	0.189	—	—	Sutkus et al. (2001)
2000	Scheduled and non-scheduled commercial aviation	214 ^d	—	677	2.9	—	—	—	—	Owen et al. (2010)
2000	Civil and military aircraft	169	—	—	2.15	—	—	—	—	Gauss et al. (2006)
	Civil aircraft	152	—	—	1.95	—	—	—	—	Gauss et al. (2006)
	Military (difference)	44	—	—	0.2	—	—	—	—	Gauss et al. (2006)
	Commercial aviation	181	224	572	2.51	0.541	0.076	0.145	—	Kim et al. (2007)
2001	Commercial aviation	170	210	536	2.35	0.464	0.063	0.136	—	Kim et al. (2007)
2002	Commercial aviation	171	211	539	2.41	0.480	0.064	0.137	—	Kim et al. (2007)
	Civil aviation	156	193	492	2.06	0.507	0.063	—	3.9	Eyers et al. (2004)
	Military aviation	19.5	24.1	61	0.178	0.647	0.066	—	—	Eyers et al. (2004)
	Civil + Military aviation	176	217	553	2.24	1.150	0.129	—	>3.9	Eyers et al. (2004)
2003	Commercial aviation	176	218	557	2.49	0.486	0.062	0.141	—	Kim et al. (2007)
2004	Commercial aviation	188	233	594	2.69	0.511	0.063	0.151	—	Kim et al. (2007)
	Commercial aviation ^e	174	215	550	2.456	0.628	0.090 ^f	0.102 ^g	6.1	Wilkerson et al. (2010)
2005	Commercial aviation	203	251	641	2.9	0.554	0.065	0.163	—	Kim et al. (2007)
2006	Commercial aviation	188	233	595	2.656	0.679	0.098 ^f	0.111 ^h	6.8	Wilkerson et al. (2010)
2008	From ICAO commercial air carriers—traffic database	229	282	725	3.21	0.688	0.092	0.183	5.7	Fuel demand by Chèze et al. (2011)
Forecasted trend										

2020	Scheduled and non-scheduled commercial aviation	336	—	1062	4	—	—	—	—	Owen et al. (2010)
2025	—	317	390	1001	4	0.951	0.127	0.253	7.9	Fuel demand forecast by Chèze et al. (2011)

Emission indices										
EI	Mean emission indices	—	1230	3160	14	3	0.4	0.8	0.025	Lee et al. (2010)
LTO cycles										
2000	Commercial aviation	12.9	15.9	40.8	0.197	—	—	0.010	—	Kim et al. (2007)
2001	Commercial aviation	12.3	15.1	38.9	0.191	—	—	0.010	—	Kim et al. (2007)
2002	Commercial aviation	12.2	15.0	38.6	0.194	—	—	0.010	—	Kim et al. (2007)
2003	Commercial aviation	12.4	15.3	39.2	0.199	—	—	0.010	—	Kim et al. (2007)
2004	Commercial aviation	12.9	15.9	40.8	0.21	—	—	0.010	—	Kim et al. (2007)
2005	Commercial aviation	13.9	17.1	43.9	0.227	—	—	0.011	—	Kim et al. (2007)

a) Type of fleet, as specified in different estimates; b) NO_x is expressed as NO₂ in Sutkus et al. (2001), Gauss et al. (2006) and Wilkerson et al. (2010); c) SO_x expressed as SO₂; d) normalized to the IEA total aviation fuel sales figure (see Owen et al. (2010)); e) corrected global fuel burn results (see Wilkerson et al. (2010)); f) HC expressed as CH₄; g) expressed as S-SO_x, assuming that 96.3% of the SO_x-S was partitioned to SO₂-S and 3.7% to S(VI)-S (particle); h) expressed as S-SO_x, assuming that 98% of the SO_x-S was partitioned to SO₂-S.

4086 **Table 3.** List of recent studies in the literature that measure EIs directly from engine or airplane tests. The table also reports studies on hydrocarbon profiles.
4087 Some information about tested aircraft and engine models, selected thrust and sampling methodologies and analytical techniques, type of fuel, date and
4088 location of experiments is also given.

Airframe/Engine	Analyzed compounds	Sampling and experimental (sampling system [analytical methods])	Tested regimes and [fuels]	References
F101 (Military TF with reheat used on the B-1B aircraft); F110 (Military TF with reheat used on the F-16C and F-16D aircraft)	CO ₂ , CO, NO _x , total hydrocarbons, individual organic species	Samples collected from each engine using a probe positioned just behind the exhaust nozzle	Four power settings from idle to intermediate power	Spicer et al. (1992)
TF-39 (Military TF of Lockheed C-5) and CFM-56 (TF)	CO, NO, NO _x , total hydrocarbons, C ₂ to C ₁₇ organics, PAHs, aldehydes	Sampling: sampling rake behind the engine. Experimental: non-dispersive infrared instruments, chemiluminescence, FID, polymeric adsorbent (XAD) and DNPH cartridges[GC/MS, GC/FID], On-Line Cryogenic Trap/GC, canister[GC/MS], Total Hydrocarbon Analyzer	Idle, 30%, 80%; [JP-4; JP-5; JP-8]	Spicer et al. (1984;1994)
PW 305 (TF in small business jets)	N ₂ O, CH ₄	Sampling: gas samples collected in the core of the engine without any bypass air. Experimental: infrared absorption spectroscopy	5.5%; 23.5%; 33.4%; 71.4%; 95.6%	Wiese et al. (1994)
Various military aircraft: T56-A-7; TF39-GE-1C ; GTCP85-180; GTCP-165-1 ; T700-GE-700; J69-T-25; J85-GE-5A; F110-GE-100; F108-CF-100 ; TF33-P-7/7A; F101-GE-102 ; TF33-P-102; F117-PW-100; AFB F118-GE-100; F404-GE-F102/400; F110-GE-129; F100-PW-100; F100-PW-229; T64-GE-100; TF34-GE-100A (All Military)	CO ₂ ; CO; NO _x ; NMHCs; Aldehydes and ketones; VOCs; filterable and condensable particulate	Sampling: various test cells, hush house exhaust rate determined using three methods: carbon balance, tracer gas and F-factor. Experimental: various US-EPA' methods, including continuous emissions monitoring system; canister [GC/MS; GC/FID]; HI-VOL [lab analysis]	Idle; Approach; Intermediate; Military; Afterburner; [JP-8]	Gerstle et al. (1999)
Research aircraft: VFW-Fokker 614 ATTAS. Engine: Rolls-Royce/SNECMA M45H Mk501 (TF)	Aerosol size distribution and chemical composition (total carbon, BC)	Sampling: ground-based measurements (also report in-flight measurements). Experimental: filter substrates[thermal technique], PCASP-100X	Different engine thrust levels: idle run and take-off	Petzold and Schröder (1998); Petzold et al. (1999)
Fighter aircraft: F-22 Raptor (Military); Engine: F119-PW-100 (TF with reheat)	CO ₂ ; CO; NO _x ; NMHCs; Filterable and condensable particulate; Aldehydes and ketones; VOCs	Sampling: engine exhaust sampling rake system; augmentor tube slipstream sampling system. Experimental: various US-EPA' methods: continuous emissions monitoring system; canister [GC/MS; GC/FID]; HI-VOL [lab analysis]	Idle (10%); approach (20%); Intermediate (70%); Military (100%); Afterburner (150%); [JP-8]	Gerstle et al. (2002)
NASA Boeing 757; Engine: RB-211-535E4 (TF)	CO ₂ , H ₂ O, HONO, HNO ₃ , SO ₂ , SO ₃ , H ₂ SO ₄ , nonmethane hydrocarbons, aerosol size, BC	Sampling: 1 m down steam of the turbine exhaust, aerosol-sampling probe was also affixed to the blast fence 25 m downstream of the engine exhaust plane. Experimental: IR spectrometer, DMA, OPC, aethalometer, grab samples, tunable diode laser, AMS	A range of power settings from idle to near take-off thrust; [JP-5, low and high S (810 and 1820 ppm S)]	EXCAVATE: Anderson et al. (2005;2006)

Jet trainer: T-38A Talon; Engine: 85-GE-5A (TJ)	CO ₂ , aerosol size, BC, nonmethane hydrocarbons, SO ₂ , CO ₂ , SO ₃ , H ₂ O, HONO, H ₂ SO ₄ , HONO, HNO ₃	Sampling: 1 m down steam of the turbine exhaust. Experimental: IR spectrometer, DMA and OPC, aethalometer, grab samples, tunable diode laser, AMS	A range of power settings from idle to near take-off thrust; [JP-5 (810 ppm S)]	EXCAVATE: Anderson et al. (2005)
Fighter: F-18 (Military). Engine: F404-GE-400 in twin-engine (TF with reheat)	Particle mass concentration, PAHs, BC	Sampling: Navy jet engine exhaust emissions from tethered aircraft, measurements at a site on the active flightline tarmac, directly from the exhausts of tethered aircraft. Experimental: DustTrak particle mass monitor, PAS, photoacoustic analyzer, Gundel denuder sampler (with PUF/XAD/PUF “sandwich” cartridges), SMPS, MOUDI cascade impactor	Power-setting increases from 65% to 70%, and from 70% to 80%	Rogers et al. (2005)
Engine: dismantled T700-GE-401 (TS), which is fitted in Seahawk, Super Cobra, and Jayhawk helicopters (Military)	Particle mass concentration, PAHs, BC	Sampling: Navy jet engine exhaust emissions from engine maintenance test cells, measurements at Aircraft Intermediate Maintenance Department facility. Experimental: DustTrak particle mass monitor, PAS, photoacoustic analyzer, Gundel denuder sampler (with PUF/XAD/PUF “sandwich” cartridges), SMPS, MOUDI cascade impactor	Power-setting increases from idle to 98%	Rogers et al. (2005)
NASA Boeing 757; Engine: RB211-535-E4 (TF)	Gaseous carbon species	Sampling: 10 m behind the engine exit plane. Experimental: Canister, analyses of whole air samples [GC/FID, GC/ECD, GC/MS]	4–7%; 26%; 47%; 61%; [JP-5 low and high S]	EXCAVATE Anderson et al. (2006)
Bell helicopter; UH-1H (TS)	22 PAHs	Sampling: engine placed in a testing chamber, exhaust samples collected from the stack of the chamber using an isokinetic sampling system. Experimental: GC/MS	Five power settings: idle (50%), fly idle (67%), beed band check (79%), inlet guide vane (95%), and take off (100%); [JP-4]	Chen et al. (2006)
Military jet fighters: F-15 Eagle and the F-16 Falcon aircraft. Engines: PW F-100-PW-100 (TF with reheat)	Automatic measurements: CO ₂ , CO, NO, NO ₂ , total hydrocarbons	Sampling: extractive sampling at 23 m behind the exhaust exit plane for tests at idle through military power, and at 38 m for afterburner tests; optical remote sensing measurements 23 m behind the engine exit plane. Experimental: automatic measurements; canisters [GC/MS]; DNPH-coated cartridges [HPLC/UV detector]; OP-FTIR; UV-DOAS	Ground idle (65–70%), low intermediate (80%), high intermediate (85%), military (91–93%) and afterburner (reheat); [JP-8+100]	Cowen et al. (2009)
Aircraft: Boeing DC-8. Engine: CFM-56-2C1 (TF)	CO, CO ₂ , NO, NO ₂ , HONO, total VOCs, gas-phase speciated hydrocarbons, particle number concentration, particle size distribution, PM _{2.5} [mass, EC/OC, SVOCs, inorganic ions, elemental composition]	Sampling: the exhaust plume was sampled at 1, 10 and 30 m downstream of the engines. Experimental: continuous and time-integrated instruments: IR absorption, TILDAS, PTR-MS, AMS, canister[GC/MS, GC/FID], DNPH cartridges[HPLC], TEOM, CPC, SMPS, DMA, PM-2.5 cyclones [47mm PTFE filter], PM-2.5 cyclones [47mm QFF+PUF], ELPI, aethalometer, PAH analyzer; lab analyses on filters and PUF [GC/MS, TOA@NIOSH, ion chromatography, XRF]	“EPA test matrix” (typical LTO); “NASA test matrix” including 11 power settings); [3 fuels: base fuel, high sulfur (1639 ppm), high aromatic]	APEX-1: Wey et al (2006) ; Knighton et al. (2007) ; Wormhoudt et al. (2007) ; Yelvington et al. (2007) ; Wong et al. (2008) ; Onash et al. (2009) ; Kinsey (2009)

Aircraft: B737-700; B737-300. Engines: CFM56-7B24, CFM56-3B1, CFM56-3B2 (all TF)	CO ₂ , gas-phase speciated hydrocarbons, particle number concentration, particle size distribution, PM _{2.5} [mass, EC/OC, SVOCs, inorganic ions, elemental composition, PAHs]	Sampling: on-wing at the ground run-up enclosure; 1, 30 and 54 m from the exhaust nozzle exit. Experimental: continuous and time-integrated instruments: IR absorption, canister[GC/MS, GC/FID], DNPH cartridges[HPLC], TEOM, CPC, SMPS, EEPS, DMA, PM-2.5 cyclones [47mm PTFE filter, 47mm QFF+PUF], ELPI, aethalometer, PAH analyzer; lab analyses on filters and PUF [GC/MS, TOA@NIOSH, ion chromatography, XRF], AMS	4%, 7%, 30%, 40%, 65%, 85%; [Jet-A]	APEX-2: Agrawal et al. (2008) ; Kinsey (2009) ; Timko et al. (2010b;c)
Aircraft: B737-300, Embraer ERJ-145, A300, B775, plus Learjet Model 25. Engines: CFM56-3B1, AE3007A1E, AE3007A1/1, PW4158, RB211-535E4-B (all TF), plus CJ610-8ATJ (TJ)	CO ₂ , gas-phase speciated hydrocarbons, particle number concentration, particle size distribution, PM _{2.5} [mass, EC/OC, SVOCs, inorganic ions, elemental composition]	Sampling: the exhaust plume was sampled at a location 1, and 30 m downstream of the engines (sometimes at 15 and 43 m); Sampling was done at the centre-line using a single probe. Experimental: continuous and time-integrated instruments: IR absorption, TILDAS, quantum cascade-TILDAS, canister[GC/MS, GC/FID], DNPH cartridges[HPLC], TEOM, CPC, SMPS, EEPS, DMA, PM-2.5 cyclones [47mm PTFE filter, 47mm QFF+PUF], ELPI, aethalometer, PAH analyzer; lab analyses on filters and PUF [GC/MS, TOA@NIOSH, ion chromatography, XRF], AMS	4%, 7%, 15%, 30%, 45%, 65%, 85%, 100% [slightly varying for some engines, see Kinsey (2009)]; [Jet-A]	APEX-3: Knighton et al. (2007) ; Kinsey (2009) ; Timko et al. (2010b;c)
Military helicopters: Blackhawk, Apache: T700-GE-700 and T700-GE-701C (TS)	CO ₂ , H ₂ O, CO, NO, and N ₂ O (FTIR); particle number, mass and size distributions, smoke number (automatic); elements, ions, EC, OC (on PM filters)	Sampling: extractive sampling at the engine nozzle, plus extractive sampling (4.14 m) and remote-sensing at a predetermined distance downstream of the engine exhaust plane. Experimental: FTIR, TDLAS, UV DOAS, OP-FTIR; CPC, DMA, SMPS, TEOM, smoke machine, sandwiched PM ₁ impaction-style sampler [XRF, ion chromatography, TOA@NIOSH]	Idle, 75%, max; [JP-8, FT]	Cheng (2009) ; Cheng et al. (2009) ; Cheng and Corporan (2010)
Military transport (cargo) aircraft: Lockheed C-130 Hercules. Engine: T56-A-15 (TP)	CO ₂ , H ₂ O, CO, NO, and N ₂ O (FTIR); particle number, mass and size distributions, smoke number (automatic); elements, ions, EC, OC (on PM filters)	Sampling: at the engine exit plane and at 5 and 15 m downstream of the engine exit. Experimental: remote sensing: FTIR, TDLAS, UV DOAS, OP-FTIR; Extractive measurements: on-line gas analyzer, cross-filter correlation spectroscopy, chemiluminescence, CPC, SMPS, TEOM, smoke machine, PM ₁ sampler [XRF, ion chromatography, carbon analyzer]	Low speed ground idle (4%); high speed ground idle (7%); flight idle (20%); cruise (41%); max (100%); [JP-8, FT]	Cheng et al. (2008) ; Corporan et al. (2008) ; Cheng (2009) ; Cheng and Corporan (2010)
Military bomber: B-52. Engine: TF33-P-3/103 (TF)	CO ₂ , H ₂ O, CO, NO, and N ₂ O (FTIR); particle number, mass and size distributions, smoke number (automatic); elements, ions, EC, OC (on PM filters)	Sampling: extractive sampling at the engine nozzle, plus extractive sampling and remote-sensing at a predetermined distance downstream of the engine exhaust plane. Experimental: FTIR, TDLAS, UV DOAS, OP-FTIR; CPC, SMPS, TEOM, smoke machine, PM ₁ sampler [XRF, ion chromatography, carbon analyzer]	TF33 (idle, 80%, 90%, 95%); [JP-8, FT]	Cheng (2009) ; Cheng and Corporan (2010)
Update and consolidation of the existing HAPs profile using data from Spicer et al. (1994) , EXCAVATE and APEXs campaigns	Hydrocarbons, EIs and profiles (mass fraction)	Data analysis	Various	Knighton et al. (2009)

Military transport (cargo) aircraft: Lockheed C-130 Hercules. Engine: Allison T56 (TP)	CO ₂ , CO, NO _x , total hydrocarbons, organic gases including carbonyls	Experimental: non-dispersive IR, cross-filter correlation spectroscopy, chemiluminescence, FID, PTR-MS, canister[GC/MS], DNPH cartridges[HPLC]	Low speed ground idle, High speed ground idle, Flight idle Cruise, Maximum power; [JP-8]	Spicer et al. (2009)
Jet fighter: F-15. Engine: PW F100-PE- 100 (TF with reheat)	CO ₂ , CO, NO _x , total hydrocarbons, organic gases including carbonyls	Experimental: non-dispersive IR, cross-filter correlation spectroscopy, chemiluminescence, FID, PTR-MS, canister[GC/MS], DNPH cartridges[HPLC]	Idle, Low intermediate, High intermediate, Military, Afterburner; [JP8+100]	Spicer et al. (2009)
Summary of the APEX1–3 campaigns: CFM56-2C1, CFM56-7B24, CFM56-3B1, CFM56-3B2, AE3007A1E, AE3007A1/1, P&W 4158, RB211-535E4-B (all TF), and CJ610-8ATJ (TJ)	Physical and chemical characterization of PM; PM mass, particle number concentrations and size, BC, surface-bound PAHs; inorganic ions, EC, OC, SVOCs, elements	As for APEX1–3 campaigns	LTO and others	Kinsey et al. (2010; 2011)
Pratt & Whitney; PW three high-bypass TF, representing two different distinct engine model types	Total particulate mass, chemical composition and size distributions of the emitted oil	Sampling: Particulate matter emitted from the lubrication system overboard breather vent with a self-designed collecting and diluting apparatus. Experimental: C-TOFAMS, TEOM, engine exhaust particle sizer, CPC and ultra high sensitivity aerosol spectrometer	Cycles from idle to 65- 70% thrust	Yu et al. (2010)
NASA DC-8; CFM56-2C1 (TF)	CO ₂ , CO, NO _x , SO ₂ , CH ₄ , N ₂ O, HONO, total and speciated hydrocarbons, hazardous air pollutants; particle measurements included number density, size distribution, mass, aerosol chemical composition, and black carbon composition	Sampling: from inlet probes positioned 1 and 30 m downstream of the aircraft's engines; aged plumes at 145 m away from the engine output in the direction of the predominant wind, 1.3 m above the ground. Experimental: NDIR, CPC, SMPS, EEPs, DMS, MAAP, PAS 2000, AMS, CCN, TILDAS, PTR-MS, conventional gas analyzers, TEOM	7 thrusts: LTO + 4%(idle); 45%(approach); 65%(cruise); [JP-8, FT (Shell), FT (Sasol)]	AAFEX: Anderson et al. (2011) , Santoni et al. (2011)
KC-135T Stratotanker (Military); CFM56- 2B1 (TF)	CO ₂ , CO, O ₂ , NO _x , total hydrocarbon; PM, particle number concentration and size (after exhausts dilution in smog chamber)	Sampling: exhaust sampled using a rake inlet installed 1 m downstream of the engine exit plane; a dilution sampler and portable smog chamber were also used. Experimental: five-gas exhaust gas analyzer; canister[GC/MS], PM _{2.5} cyclone[QFF and PTFE filters, Tenax TA sorbent, GC/MS, OC/EC analyzer], SMPS, AMS	4%, 7%, 30%, 85%; [JP- 8]	Presto et al. (2011) ; Miracolo et al. (2011)

Helicopters; Allison T63-A-700 (TS)	CO ₂ , CO, NO _x , CH ₄ , and C ₂ H ₄ , unburned hydrocarbons, number and size of particles, BC	Samples were extracted from the engine exit plane via temperature-controlled probes, charcoal tubes, DNPH tubes; NDIR, FTIR, FID, CPC, SMPS, MAAP, GC/MS	3% (low-speed idle), 7% (high-speed idle), 15% (intermediate), 85% (cruise); [JP-8, a synthetic paraffinic kerosene, and four two-component surrogate mixtures]	Cain et al. (2013)
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Used acronyms: AMS=aerosol mass spectrometer; BAM=beta-attenuation mass monitor; CPC=condensation particle counter; C-TOF AMS=time-of-flight aerosol mass spectrometer; DMA=differential mobility analyser; EEPS=engine exhaust particle sizer; ELPI=electrical low pressure impactor; FTIR=Fourier transform infrared spectroscopy; GC/ECD=gas chromatography/electron capture detector; GC/FID=gas chromatography/flame ionization detector; GC/MS=gas chromatography/mass spectrometry; HI-VOL=high volume PM sampler; LIDAR=laser interferometry detection and ranging; MAAP=multi-angle absorption photometer ; NDIR=non-dispersive infrared spectroscopy; OPC=optical particle counting and photometry; OP-FTIR=open-path Fourier transform infrared spectroscopy; PAS=photoelectric aerosol sensor; PTFE=Teflon; PTR-MS=proton-transfer reaction mass spectrometry; QFF=quartz fibre filter; SEM/EDX=scanning electron microscopy/energy-dispersive X-ray spectroscopy; SMPS=scanning mobility particle sizer spectrometer; TDLAS=tunable diode laser absorption spectroscopy; TEOM=tapered element oscillating microbalance; TF=turbofan; TILDAS=tunable infrared differential absorption spectroscopy; TJ=turbojet; TOA=thermo-optical OC-EC analyzer (@used method); TP=turpoprop; TS=turboshaft; UV-DOAS=UV differential optical absorption spectroscopy; VOC=volatile organic compounds; XRF=X-ray fluorescence spectroscopy.

4103 **Table 4.** List of recent studies available in the literature reporting EIs during real aircraft operation. The table also reports supplementary information (if
4104 available) about the target of the study, period and location of experiments, tested aircraft or engine models, measured pollutants, analysed LTO phases and
4105 sampling methodologies. The list of acronyms is provided in Table 3.

Target; Period; Airport	Analyzed compounds	Sampling; Analytical	Engine thrusts (if know) or LTO phases	References
In service military and civil aircraft at various airports	CO ₂ , H ₂ O, CO, NO, N ₂ O	Measurements performed at distances of 20-40 m to the nozzle exit perpendicular to the exhaust flow via ground-based FTIR analysis	Various thrusts	Heland and Schafer (1997;1998)
Various (90) in service aircraft: from gulfstream executive jets to Boeing 747-400s at London Heathrow Airport (UK)	CO ₂ , CO, NO, hydrocarbons	The remote sensor positioned at ground level. Experimental: non-dispersive IR spectroscopy, dispersive UV spectrometer	Mix of idle, taxi-out and take-off modes	Popp et al. (1999)
Emission indices of different aircraft engines using non-intrusive measurements at Frankfurt/Main (GER), London-Heathrow (UK), Vienna (AT) airports	CO ₂ , CO, NO, NO ₂ , ethene, ethine, formaldehyde	Open paths of 80 up to 150 m length were installed in parallel directly behind the aircraft. Experimental: FTIR with MIDAC spectrometer, FTIR with K300 spectrometer, DOAS	Aircraft operating conditions, idling aircraft	Schäfer et al. (2003)
30 individual planes, ranging from TP to jumbo jets; August 2001; J.F. Kennedy Airport (USA)	CO ₂ , NO, NO ₂	Measurements within 350 m of a taxiway and 550 m of a runway. Experimental: automatic (IR), TILDAS	Taxiway thrust and take-offs	Herndon et al. (2004)
In-use commercial aircraft; period: 2001-2003; Airports: J.F. Kennedy airport in New York City and Logan airport in Boston (USA)	Particulate matter, number concentration and size distributions	Extractive sampling of the advected plumes of aircraft using a novel approach, 200 m of an active taxiway and runway. Experimental: ELPI, CPC	Several different types of plumes were sampled, including approach (landing) and engine start-up in addition to idle, taxi, and take-off	Herndon et al. (2005)
45 intercepted plumes identified as being associated with specific aircraft: regional jets, B737s, MD88s, and B757s; Period: May 2003; Logan airport in Boston (USA)	CO ₂ ; Formaldehyde, acetaldehyde, benzene, and toluene, as well as other hydrocarbon species; NO _y	Ambient air is continuously analyzed through a sample port located near the roof on the front of the truck. Experimental: IR, PTR-MS; TILDAS; total reactive nitrogen instrument	Idle, taxi, approach (or landing), and take-off, as well as engine-start modes	Herndon et al. (2006)
Real time data at Los Angeles International Airport (USA); Period: September 23-29, 2005	UFPs (diameter <100 nm), black carbon, PM _{2.5} mass, and chemical species (PAHs, butadiene, benzene, acrolein, formaldehyde)	At blast fence (140 m from the take-off) and five downwind sites up to 600 m from the take-off runway. Experimental: SMPS (DMA/CPC), aethalometers, E-BAM, automatic PAHs analyzer, canister, cartridge	—	Fanning et al. (2007); Zhu et al. (2011)
Impact of airport emissions at Zurich–Kloten airport (Switzerland); Period: June 2004 to July 2004	NO, NO ₂ , CO, CO ₂ , VOCs	Measurements with in-situ and open-path devices; COV samples taken directly within the plume of the engine, about 50–100m behind an aircraft, at a height of 1m. Experimental: FTIR; DOAS; canister [GC/FID]	—	Schürmann et al. (2007)

Emissions from in-use commercial aircraft engines analyzed using continuous extractive sampling and associated with specific engine using tail numbers; Period: September 2004; Location: Hartsfield-Jackson Atlanta International Airport (USA)	CO ₂ , CO, NO, NO ₂ , formaldehyde, particle number, BC, particle size, mass-based composition	Two mobile laboratories located downwind of active runways. Experimental: Automatic (IR); TILDAS; CPC; MAAP; SMPS; DMS; AMS	Various	JETS/APEX-2 campaign: Herndon et al. (2008)
Plume characterization from commercial aircraft at Brisbane Airport (AUS)	CO ₂ , SO ₂ , NO _x , particle mass, number concentration and size	Plume capture and analysis system mounted in a four-wheel drive vehicle positioned in the airfield 60 to 180 m downwind of aircraft operations. Experimental: CPC, SMPS, NO _x analyzer, aerosol photometer fitted with a PM _{2.5} impactor	Normal airport operations, taxiing phase	Johnson et al. (2008)
In-use commercial airfreight and general aviation at Oakland International Airport (USA); Period: August 20-29, 2005;	Formaldehyde, acetaldehyde, ethene, propene, and benzene	At the end of an active taxiway next to the main runway. Data collected on an ambient sampling manifold consisting of a 3.8 cm diameter tube, ~7 m long drawing ~150 slpm. Experimental: TILDAS; proton transfer reaction mass spectrometer measurements	Idle (taxiway/runway)	JETS/APEX-2 campaign: Herndon et al. (2009)
Real world conditions, 280 individual aircraft at Brisbane Airport (AUS)	Particle number concentration, size and mass (PM _{2.5}), CO ₂ , NO _x	80 m from the aircraft using a novel mobile measurement system. Experimental: CPC, SMPS, NO _x analyzer, aerosol photometer fitted with a PM _{2.5} impactor	Various modes of LTO cycles including idle, taxi, landing, and take-off	Mazaheri et al. (2009)
In-use commercial aircraft at Chicago Midway Airport and O'Hare International Airport (USA); Period: February 2010	CO, NO, NO _x , oil leaks	Mobile laboratory located at downwind locations to monitor air advected from the active taxiways (30–150 m). Experimental: TILDAS; HR-ToF AMS; MAAP, CPC	—	Yu et al. (2012)
Emission of Roanoke Regional Airport in Virginia (USA); Period: July 2011 - February 2012	CO ₂ , NO _x , particle number, BC	A mobile eddy covariance laboratory with a mast extending nearly 15 m above ground level and placed near active runways. Experimental: automatic devices, CPC, aethalometer	Idle/taxi and take-off	Klapmeyer and Marr (2012)
Real-time measurements of aircraft engine specific emissions at Oakland International Airport (USA); Period: August 26, 2005	CO ₂ , particle number concentration, size distributions, PM mass	100-300 m downwind of an active taxi-/runway. Experimental: Automatic IR, Cambustion DMS500, CPC, SMPS, MAAP	Normal LTO operations	Lobo et al. (2012)

4108 **Table 5.** List of recent studies available in the literature conducted at airports or in their surroundings. The table also reports supplementary information (if
4109 available) about the target of the study, period and location of experiments, tested aircraft or engine models, measured pollutants, analysed LTO phases and
4110 sampling methodologies. The list of acronyms is provided in Table 3.

Target; Period; Airport	Analyzed compounds	Sampling; Analytical	Engine thrusts (if know) or LTO phases	References
Air quality data in the vicinity of Hong Kong International Airport (1997-1998) and Los Angeles International Airport (2000-2001)	CO, NO _x , SO ₂ , and respirable suspended particles	Data from routine air quality monitoring site and special study	—	Yu et al. (2004)
Airport traffic at Heathrow (UK); Period: Jul. 2001–Dec. 2004	NO _x , NO ₂	LHR2 site at 180 m north of the northern runway centreline. Experimental: Common automatic devices	—	Carslaw et al. (2006)
Ambient air and personal at Fiumicino Airport, Rome (Italy); Period: January-February 2005	23 PAHs, urinary 1-hydroxy-pyrene, micronucleus assay, Comet assay, Sister chromatid exchange	Air samples collected from airport apron, airport building and terminal/office area during 5 working days, plus a biomarker of exposure following 5 working day. Experimental: Active ECHO PUF sampler at 35 L/min for the first 20 min and at 120 L/min for the remaining 23 h and 40 min on each day, [GC/MS analysis]	—	Cavallo et al. (2006)
Individual plumes from 29 commonly used engines; Period: October 19-November 15, 2005; Location: London Heathrow (UK)	NO _x	180 m from the runway. Experimental: chemiluminescence monitor	—	Carslaw et al. (2008)
Analysis of the extent of Los Angeles International Airport emissions on downwind ambient air in a mixed use neighborhood that includes residences. Period: spring of 2003	UFP, BC, NO _x , particle-phase PAHs	Data collected at various sites in and around the airport: 500 m upwind of the north runway and downwind of the airport (500 m north and east of the centerline of the north runway; 100 m downwind of the taxiway; 100 m downwind of the south runway; 900 m downwind of the south runway) . Experimental: CPC, SMPS, DMA, aethalometer, photoelectric aerosol sensor, NO _x analyzer	—	Westerdahl et al. (2008)
APEX2-3: Oakland International Airport in August 2005, and Cleveland Hopkins International Airport in Oct-Nov 2005.	NO _x and NO _y , including HONO	Panel truck. Experimental: TILDAS; quantum cascade-TILDAS; chemiluminescence analyzer	—	Wood et al. (2008b)
Airport traffic at Warwick, Rhode Island (USA); Period: July 2005-September 2006	BC	Five monitoring sites: 4 close and 1 approx 3.7 km from the airport. Experimental: Continuous with aethalometers	—	Dodson et al. (2009)
General aviation and private jets at Santa Monica Airport (USA); Period: Spring and summer 2008	UFP, PM2.5, BC, particle bound PAHs, CO, NO _x , NO, NO ₂	Downwind of the airport using an electric vehicle mobile platform equipped with fast response instruments. Experimental: CPC,	Idle/taxi and take-off	Hu et al. (2009)

		FMPS, aethalometer, PAS, automatic measurements of gases		
Airport traffic at El Prat, Barcelona (Spain); Period: October 17-November 16, 2007	PM10, PM2.5 and PM1 continuously; PM10 (EC, OC, SO42-, NO3-, Cl-, NH4+, Al, Ca, K, Mg, Fe, S, Na, As, Ba, Bi, Cd, Ce, Co, Cr, Cs, Cu, Ga, Hf, La, Li, Mn, Mo, Nb, Ni, P, Pb, Rb, Sb, Sc, Se, Sn, Sr, Th, Ti, Tl, U, V, W, Y, Zn, Zr)	Mobile laboratory van at about 130 m from the major runway. Experimental: PM ₁₀ , PM _{2.5} and PM1 with laser-spectrometer dust monitors and PM10 on QFF using HI-VOL sampler	Take-off, sometimes landing	Amato et al. (2010)
Commercial aircraft; Period: 10–20 May 2005; Airports: Manchester and London Heathrow (UK)	Dispersion of exhaust plumes	Rapid-scanning LIDAR system installed at ground 200–330 m on the sides of runways	All modes were observed: taxiing, take-off, rotation, climb-out, approach, and landing. Landing tyre smoke	Bennett et al. (2010) ; Bennett and Christie (2011)
Commercial airliners at London Heathrow (UK): A320 232; B757 236; B747 436)	PM elemental composition, particle size spectrum	Samples of dust from the undercarriage. Experimental: SEM/EDX; aerosizer/aerodisperser	—	Bennett et al. (2011)
Ambient air and personal at the Teterboro Airport, New York/New Jersey metropolitan area (USA); Period: Summer 2006 and winter 2006–2007;	BTEX	At 15 households located close to the airport (indoor, outdoor, and personal), at the end of airport runways and an out-of-neighborhood location. Experimental: Passive samplers (48 h) [GC/MS]	—	Jung et al. (2011)
High-resolution monitoring and flight activity data to quantify contributions from LTO at T.F. Green Airport in Warwick (USA). Period: 2007–2008	Particle number concentration	Four stationary monitoring sites around the airport. Experimental: CPC	Various LTO phases, especially departures	Hsu et al. (2012)
Aircraft emissions and local air quality impacts from take-off activities at Los Angeles International Airport (USA). Periods: September 2005; Feb–Mar 2006; May 2006	Particle number concentrations and size distributions, and time integrated black carbon, PM _{2.5} mass, and chemical species	Data collected at the blast fence (~140 m from the take-off position) and 5 sites located downwind, up to 600 m from the take-off runway and upwind of a freeway. Experimental: CPC, SMPS, aethalometers, BAM, PAH Tisch Sampler, canister and cartridge samplers[lab analysis]	Taxi-way and take-off operations	Zhu et al. (2011)
Contributions of aircraft arrivals and departures to UFP at Los Angeles International Airport (USA). Period: summer 2008	Particle number concentration	Five sites around the airport. Experimental: Fast Mobility Particle Sizer	LTO phases: aircraft arrivals and departures	Hsu et al. (2013)

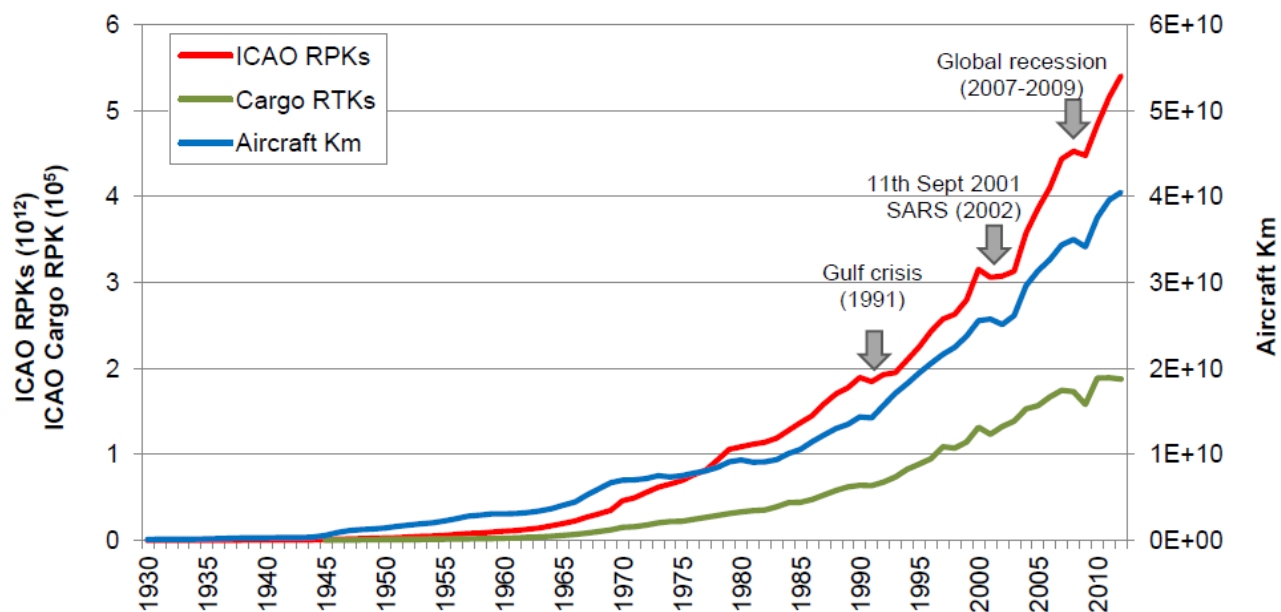


Figure 1. Absolute growth of aviation (1930–2012) recorded by ICAO in terms of RPK, RTK and aircraft kilometres. Data refers to ICAO (2013) and were taken from Airlines for America (2013).

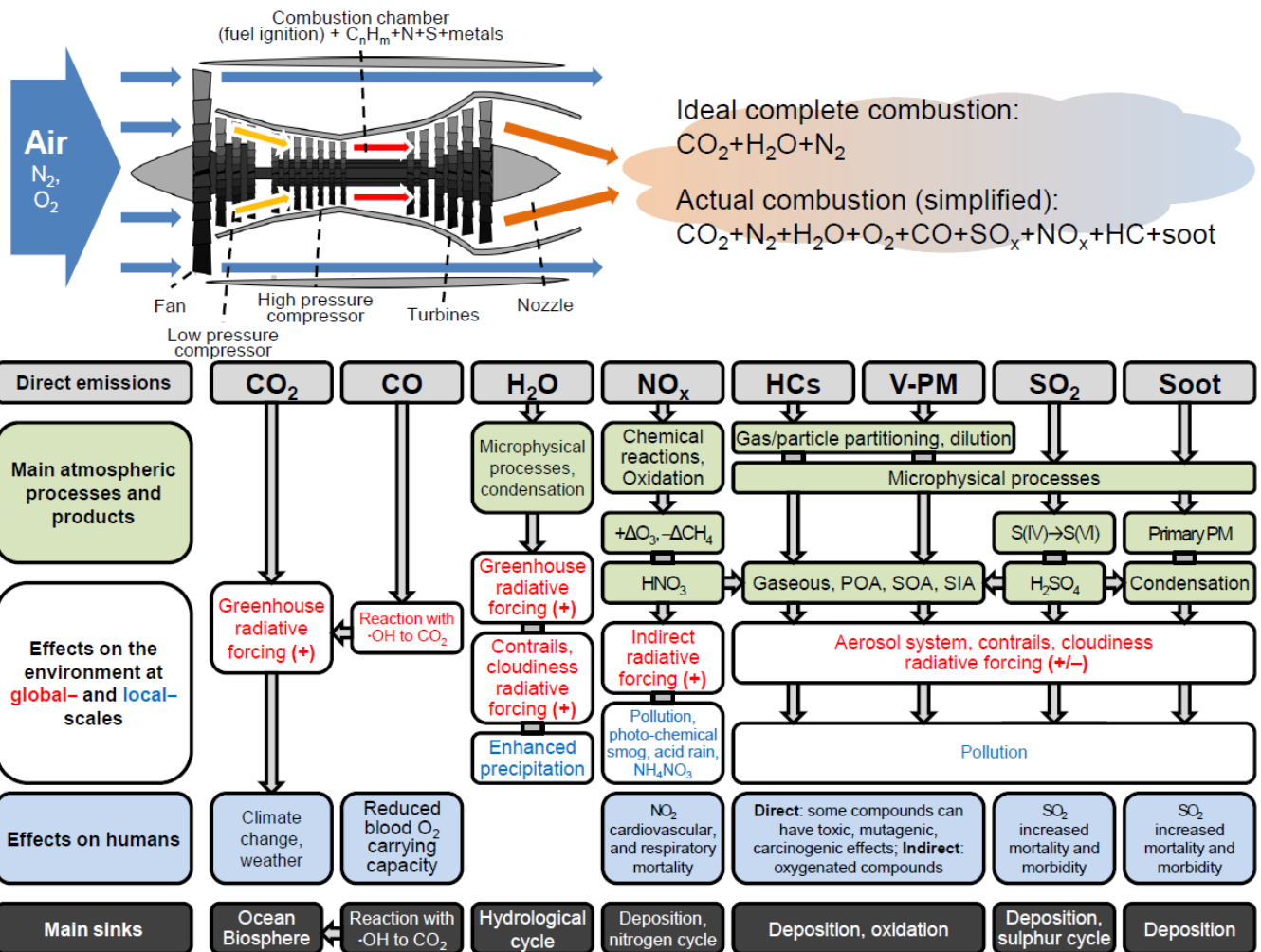


Figure 2. Simplified diagram of a turbofan engine (upper left); products of ideal and actual combustion in an aircraft engine (upper right); and related atmospheric processes, products, environmental effects, human health effects and sinks of emitted compounds (bottom). Adapted from Prather et al. (1999), Wuebbles et al. (2007) and Lee et al. (2009).

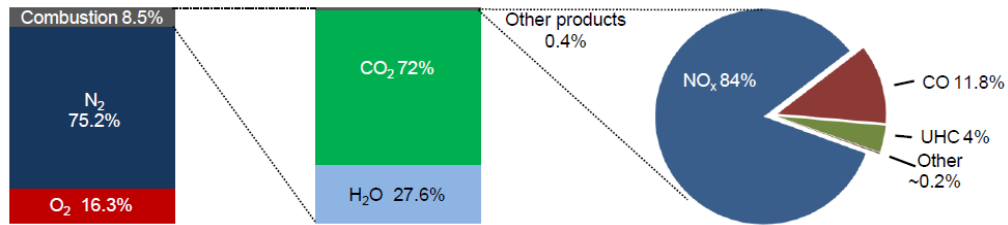


Figure 3. Division of the combustion products from an aircraft engine, adapted from Lewis et al. (1999).

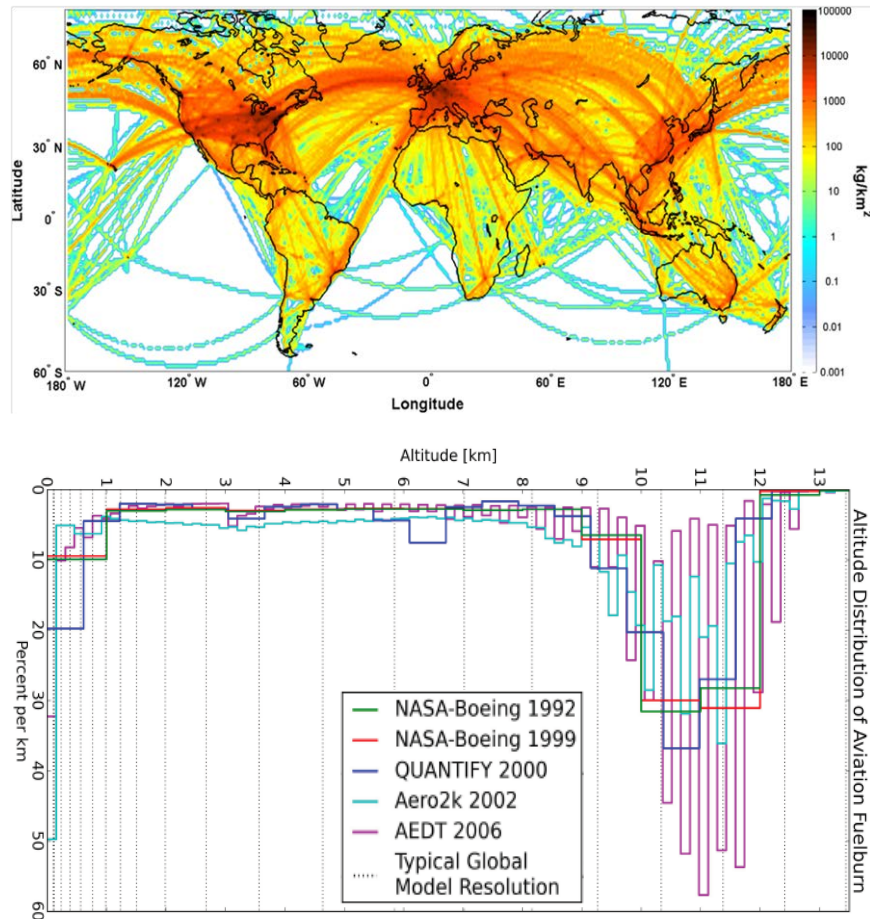


Figure 4a and 4b. Geographical and vertical distributions of aviation: a) column sum of global fuel burn from scheduled civil aviation in 2005, as reported by Simone et al. (2013) using AEIC model (Stettler et al., 2011); b) annual global vertical distribution of commercial aviation fuel burn for the NASA-Boeing 1992 and 1999 (Baughcum et al., 1996a;b; Sutkus et al., 2001), QUANTIFY 2000 (Owen et al., 2010), AERO2k (Eyers et al., 2004) and AEDT 2006 (Roof et al., 2007) datasets, taken from Olsen et al. (2013).

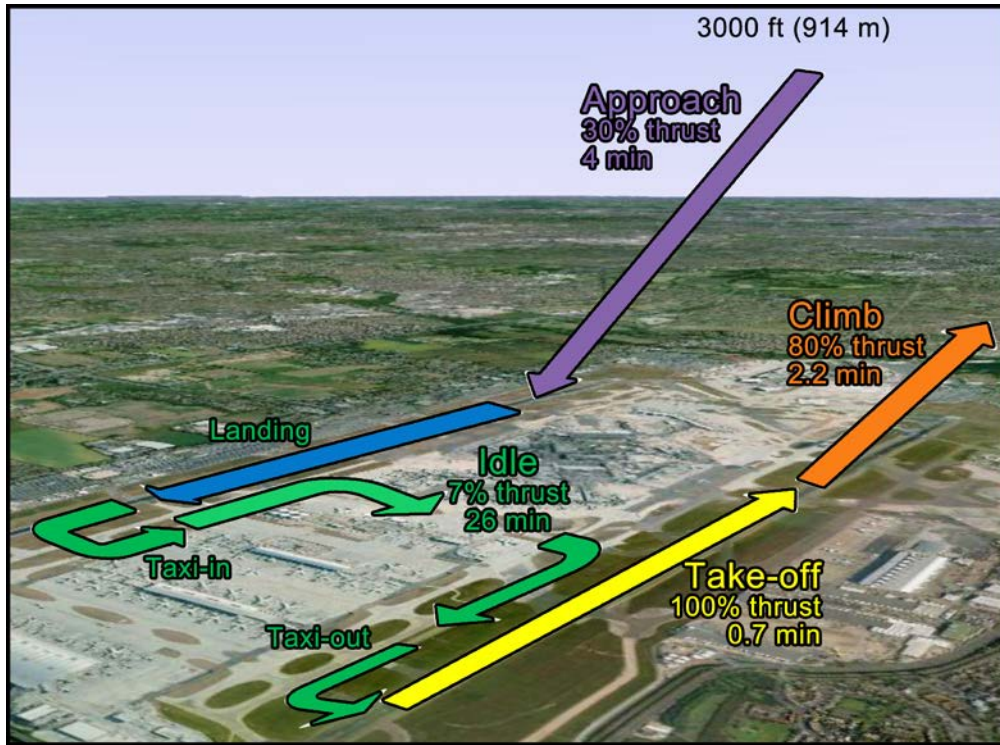


Figure 5. Standard ICAO LTO cycle. Adapted from ICAO (2011).

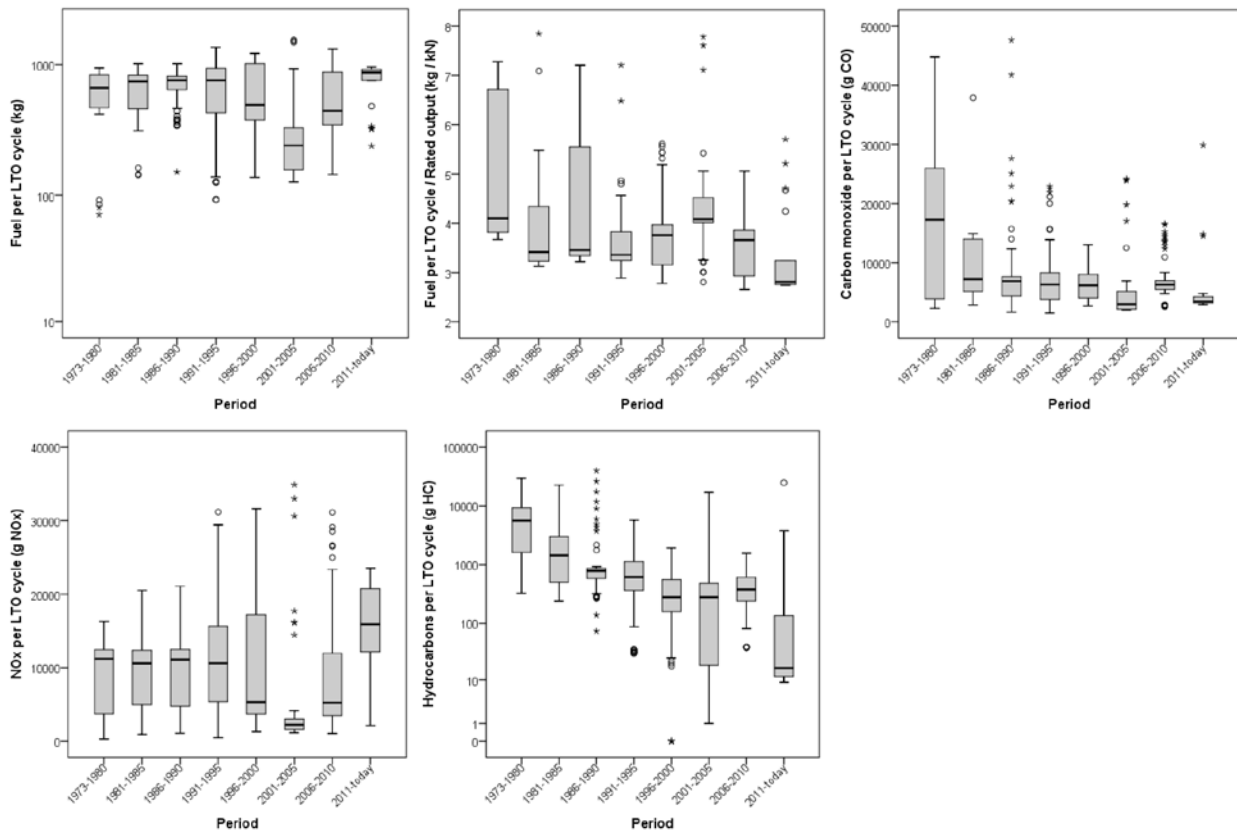
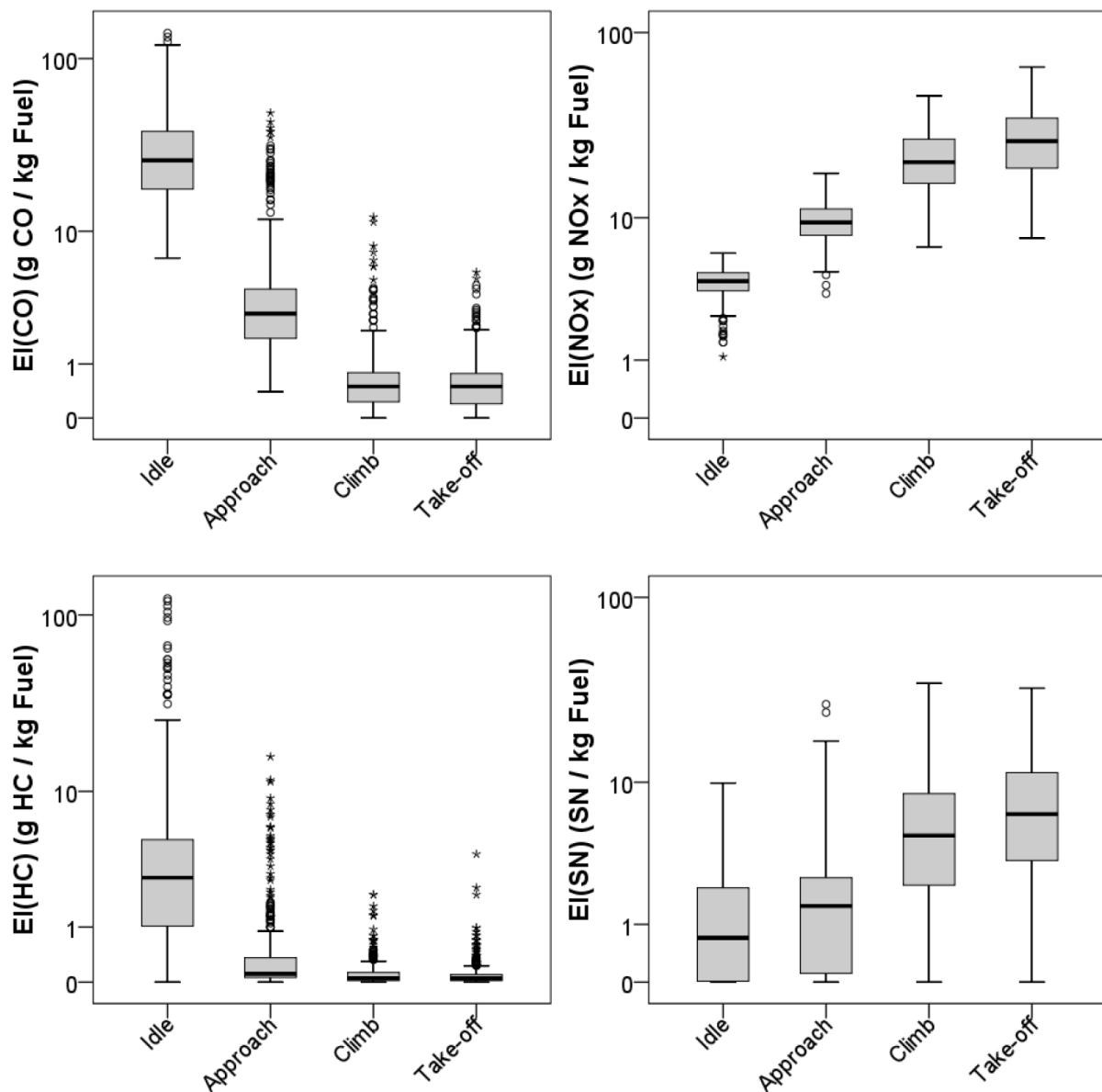


Figure 6. Burned fuel and emissions for complete standardised LTO cycle. Data from ICAO databank at April 2013 (EASA, 2013). All engines certified in each period were included in the statistics, without distinction of type, manufacturer, model or technology.



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4180 **Figure 7.** EIs provided by the ICAO databank (EASA, 2013). All in-use engines certified from
 4181 1976 to today (April 2013) are included.
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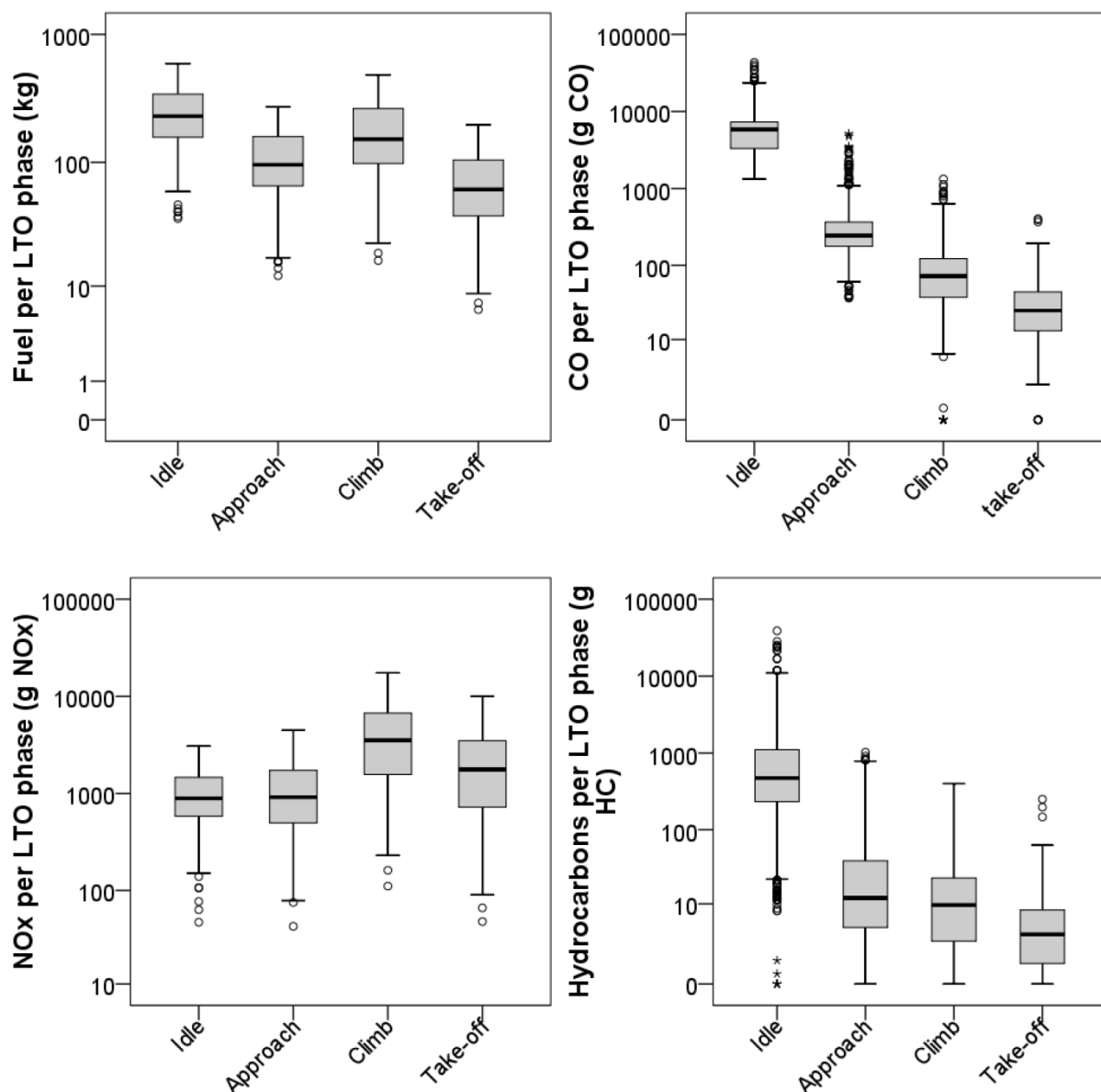


Figure 8. Fuel burned and emissions of CO, NO_x and total unburned hydrocarbons during the four LTO phases. Data were calculated from the EIs and fuel consumption provided by the ICAO databank (EASA, 2013). All in-use engines certified from 1976 to today (April 2013) were included and reprocessed as a function of LTO stages and standard times (i.e., 0.7 min for take-off, 2.2 min for climb-out, 4 min for approach and 26 min for idle).

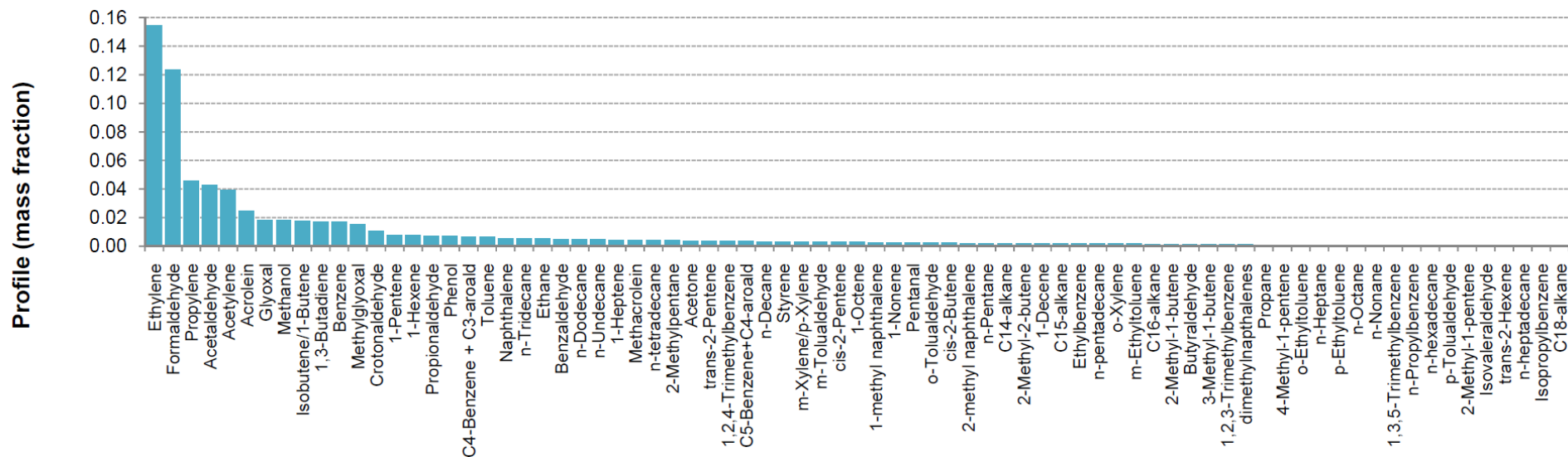


Figure 9. Results of the APEX campaigns. Profile (mass fractions) of individual hydrocarbon species. The single compounds are ordered to show decreasing fractions.